UNCLASSIFIED

| AD NUMBER |
| --- |
| **AD824724** |
| LIMITATION CHANGES |
| TO:<br>**Approved for public release; distribution is unlimited.** |
| FROM:<br>**Distribution authorized to U.S. Gov't. agencies and their contractors; Critical Technology; SEP 1967. Other requests shall be referred to Air Force Materials Laboratory, Attn: MAAE, Wright-Patterson AFB, OH 45433. This document contains export-controlled technical data.** |
| AUTHORITY |
| **AFML ltr, 12 Jan 1972** |

THIS PAGE IS UNCLASSIFIED

AFML-TR-67-273

AD 824724

# FABRICATION AND TEST OF HIGH-TEMPERATURE HONEYCOMB CORE SANDWICH CONSTRUCTION

G. WINTERMUTE
GOODYEAR AEROSPACE CORPORATION
ARIZONA DIVISION

TECHNICAL REPORT AFML-TR-67-273
SEPTEMBER 1967

This document is subject to special export controls and each transmittal to foreign governments or foreign nationals may be made only with prior approval of the Air Force Materials Laboratory, MAAE, Wright-Patterson Air Force Base, Ohio 45433.

AIR FORCE MATERIALS LABORATORY
RESEARCH AND TECHNOLOGY DIVISION
AIR FORCE SYSTEMS COMMAND
WRIGHT-PATTERSON AIR FORCE BASE, OHIO

50th Anniversary
1917 1967
AIR FORCE MATERIALS LABORATORY
Materials Research in the Air Force

ACCESSION for

CFSTI WHITE SECTION ☐

DDC BUFF SECTION ☑

UNANNOUNCED ☐

JUSTIFICATION ..................

..................

BY ..................

DISTRIBUTION/AVAILABILITY CODES

| DIST. | AVAIL. and/or SPECIAL |
|---|---|
| 2 | |

# NOTICES

# FABRICATION AND TEST OF HIGH-TEMPERATURE HONEYCOMB CORE SANDWICH CONSTRUCTION

G. Wintermute

## FOREWORD

This document is the summary technical report prepared by Goodyear Aerospace Corporation, Litchfield Park, Arizona. The work was performed under U.S. Air Force Contract No. AF33(615)-5131, "Fabrication and Test of High-Temperature Honeycomb Core Sandwich Construction."

The work was administered under U.S. Air Force Materials Laboratory, Materials Engineering Branch, Materials Application Division, U.S. Air Force Systems Command, Wright-Patterson AFB, Ohio. The project engineer for this contract was Mr. Weldon Scardino. This contract was assigned Project No. 7381, "Materials Application, " Task 738101, "Exploratory Design and Prototype Development.

This report summarizes the work accomplished and results obtained during the period 1 June 1966 through 30 June 1967.

Goodyear Aerospace has assigned the following secondary number to this report: GERA-1297.

This report was submitted in September 1967.

This report has been reviewed and is approved.

Albert Olevitch
ALBERT OLEVITCH, Chief
Materials Engineering Branch
Materials Application Division

# ABSTRACT

The primary objective of this program was the determination of the engineering properties of polybenzimidazole and polyimide honeycomb core sandwich constructions after aging at various temperatures.

The work was conducted in three phases:

I. Processing and Materials Study

II. Heat Aging and Testing

III. Data Correlation

Both press and autoclave methods were evaluated for the fabrication of sandwich face sheets and honeycomb core sandwich composites. A "pressure-point" cure technique was developed which provided a controlled, efficient processing method for preparing polyimide and PBI laminates and sandwich composites.

Technology was developed for fabricating sandwich composites by secondary bond, single stage cure, and multiple stage cure methods. The single stage cure method, using the pressure-point technique, was selected for preparing the heat aging test specimens for both the polyimide and the PBI resin systems.

Heat aging studies were conducted at 400F, 500F, and 600F. Polyimide sandwich composites exhibited excellent heat resistance at all test temperatures. The polyimide composites retained over 50% of their room temperature flexural shear strength values after 1400 hours heat aging at 400F.

The PBI sandwich composites were susceptible to oxygen degradation during fabrication and during the heat aging study. Specimens heat aged at 600F exhibited complete failure within a 300-hour period. Oxygen attack was less at lower temperatures and PBI test specimens still retained over 50% of their room temperature strength values after 600 hours aging at 400F.

# TABLE OF CONTENTS

## TABLE OF CONTENTS (contd)

## LIST OF ILLUSTRATIONS

List Of Illustrations (contd)

List Of Illustrations (contd)

# LIST OF TABLES

## LIST OF TABLES (contd)

## LIST OF TABLES (contd)

# SECTION I

## INTRODUCTION

As the overall performance of aerospace vehicles has increased, the demand for improved plastic components has also increased.

Aerospace companies active in the design of future aerospace vehicles have predicted that plastics will be used in ever increasing amounts and in more critical and demanding areas. New aircraft designs bear out this prediction.

For example, the design of the supersonic transport has approximately 13,000 square feet of plastic sandwich construction in the exterior surface areas of the airframe. These areas include wing panels, wheel-well doors, hatch covers, sections of the empennage, and portions of the nose and cabin.

The use of plastic composites in aerospace vehicle structures is desirable for a number of reasons which include:

1. High strength-to-weight ratio.

2. Resistance to vibration fatigue.

3. Corrosion resistance.

4. Thermal insulation.

5. Acoustical insulation.

The use of plastic composites for structural components on aerospace vehicles requires a knowledge of the performance of the plastic composites in the demanding environments of the aerospace craft. During sustained flight at airspeeds approaching Mach 3, skin temperatures will be at 450F to 500F for extended periods of time and will possibly reach 600F for short periods.

Throughout the past decade, research and development activity (much of it generated by the Air Force Materials Laboratory) in the area of high-heat resistant plastics has produced several thermally stable organic polymers that have excited much interest in the aerospace field. These polymers have indicated a possibility of providing structural plastic components that could perform for extended periods of time at 500F temperature or higher.

Several of these heat resistant resin systems have become available in sufficient quantity to permit extensive evaluation of physical properties and development of processing techniques.

Among these heat resistant resins, the polyimides (PI) and the polybenzimidazoles (PBI) have shown exceptional ability to retain physical properties under long time heat aging conditions. This resistance to heat degradation makes the PBI and PI resins prime candidates for use in plastic structural components on supersonic aerospace vehicles.

This program provided for the study and evaluation of PI and PBI resin systems in the form of honeycomb core sandwich composites. In a sandwich configuration, the reinforced plastic materials attain their ultimate in strength-to-weight ratio. Sandwich composites are, therefore, of increasing importance to the aerospace industry.

Processing techniques were studied and evaluated in order to select the optimum method for fabricating sandwich constructions for heat aging tests. The PBI and PI resins were more difficult to fabricate than the standard structural resins currently in general use in the aerospace industry.

The PBI and PI resins possessed a relatively high volatile release during cure. This volatile release coupled with a marked increase in resin fluidity just prior to gellation could lead to laminates that were resin starved due to excessive resin squeeze out or were quite porous due to entrapped volatiles.

Precisely controlled fabricating techniques were required in order to produce dense optimum laminates.

Heat age test panels were fabricated by a single stage autoclave method which produced superior sandwich constructions.

Heat aging tests were conducted at 400, 500, and 600F temperature levels for periods of 600 hours. Some tests were continued for 1400 hours.

## SECTION II

## SUMMARY


The polyimide resin used in this program was Skybond 700. The polybenzimidazole resin was AFR-151 (PBI). The reinforcement used in the face sheets and the honeycomb core was E-glass. The core cell size was 3/16-inch; core thickness was 1/2-inch. Two core densities - 4.0 lb and 8.0 lb. per cubic foot - were evaluated. The polyimide adhesive consisted of a modified Skybond 700 resin formulation on a glass cloth carrier. The PBI adhesive was a modified AFR-100 (PBI) resin formulation on a glass cloth carrier. The AFR-100 (PBI) resin was used for the adhesive when it was discovered that the AFR-151 (PBI) resin could not be formulated to produce a satisfactory supported adhesive film.

Processing studies were directed toward achieving optimum honeycomb core sandwich constructions for maximum heat-age performance. Sandwich composites were prepared using a secondary bond method and a single-step process. Both methods produced excellent composites.

In the secondary bond method, pre-cured face sheets were bonded to the core using an adhesive film. Both press and autoclave processing were evaluated for curing the thin four-ply face sheet laminates and for bonding the face sheets to the core. Acceptable sandwich composites were prepared using any combination of press or autoclave fabricated face sheets and press or autoclave cured secondary bonds.

The single-step process proved to be the most effective and practical technique for fabricating sandwich composites. In this method, the component parts of the sandwich construction were assembled, enclosed in a sealed bag, and subjected to heat and pressure in an autoclave. Curing of the face sheet laminates and bonding of the face sheets to the core occurred simultaneously. Processing factors studied included: cure time and temperature; pressure; pressure application; post cure; and volatile venting.

By exercise of careful control, all processing techniques produced excellent composites. However, the single stage method was chosen because it was more production oriented.

The processing studies resulted in the development of the "pressure-point" cure technique. In this technique the part was heated up under contact pressure at a controlled rate. The temperature of the part was read accurately. At a specified part temperature, pressure was applied. The pressuring point was critical. Pressure had to be applied during the conversion of the resin, after most of the volatile reaction products had been discharged, but before advanced gelation had occurred. The cure was then continued under pressure. The advantage of the pressure-point technique was an efficient, controlled, reproducible cure cycle which produced a dense, strong part.

Heat age testing specimens were prepared by the single-step process using the "pressure-point" cure technique. The testing program consisted of flatwise tensile, compressive, and flexural shear and modulus in the ribbon and transverse directions. The aging temperatures were 400F, 500F, and 600F. Complete heat age results were obtained over a test period of 600 hours. Additional flexural shear and modulus values (ribbon direction) were obtained for periods as long as 1400 hours.

The polyimide sandwich composites exhibited excellent heat resistant properties. Flexural shear strengths were still above 50% of their room temperature values after 1400 hours aging at 400F and 500F.

The PBI sandwich composites were somewhat sensitive to oxygen attack. The AFR(151) resin was seriously degraded by oxidation of the polymer chain. The inherent porosity of the sandwich face laminates and the extreme porosity of the honeycomb core presented excessive surface area which created a deleterious effect on thermal aging of the sandwich specimens. Degradation increased as aging temperature increased. At an aging temperature of 600F, complete failure occurred in less than 300 hours.

## SECTION III

## PROCESS STUDY - POLYIMIDE FACE SHEETS

### A. GENERAL

The resin used in this project was Skybond 700, a polyimide resin manufactured by the Monsanto Company. The reinforcement was 181 style E-glass with A-1100 finish. The resin and fiberglass reinforcement were combined by the Narmco Material Division, Whittaker Corp., a commercial pre-impregnator, and procured under the following designation: "Skybond 700 Prepreg 181 Cloth - #1830". Both the press method and the autoclave method were evaluated for fabricating polyimide face sheet laminates.

In the press method the prepreg plies were positioned between aluminum caul plates and subjected to heat and pressure in a heated platen press.

In the autoclave method the prepreg plies were positioned on an aluminum base plate and covered with bleeder cloth to permit release of volatile components. This lay-up was encased in a sealed bag which was attached to a vacuum system. The entire assembly was placed in an autoclave and subjected to heat and pressure.

Figure 1 shows lay-ups prepared for processing. An autoclave lay-up is on the left, a press lay-up on the right.

The face sheet laminates were a four-ply construction which produced a cured face sheet thickness of approximately .040-inch. This thickness was felt to provide good handling and processing characteristics and was sufficiently strong to assure that maximum properties of the core would be developed in the sandwich constructions.

### B. PRESS FABRICATION OF POLYIMIDE FACE SHEETS

This process study included an investigation of the following factors:

1. Application of pressure
2. Pressure.
3. Temperature profile.
4. Volatile release.
5. Cure cycle.

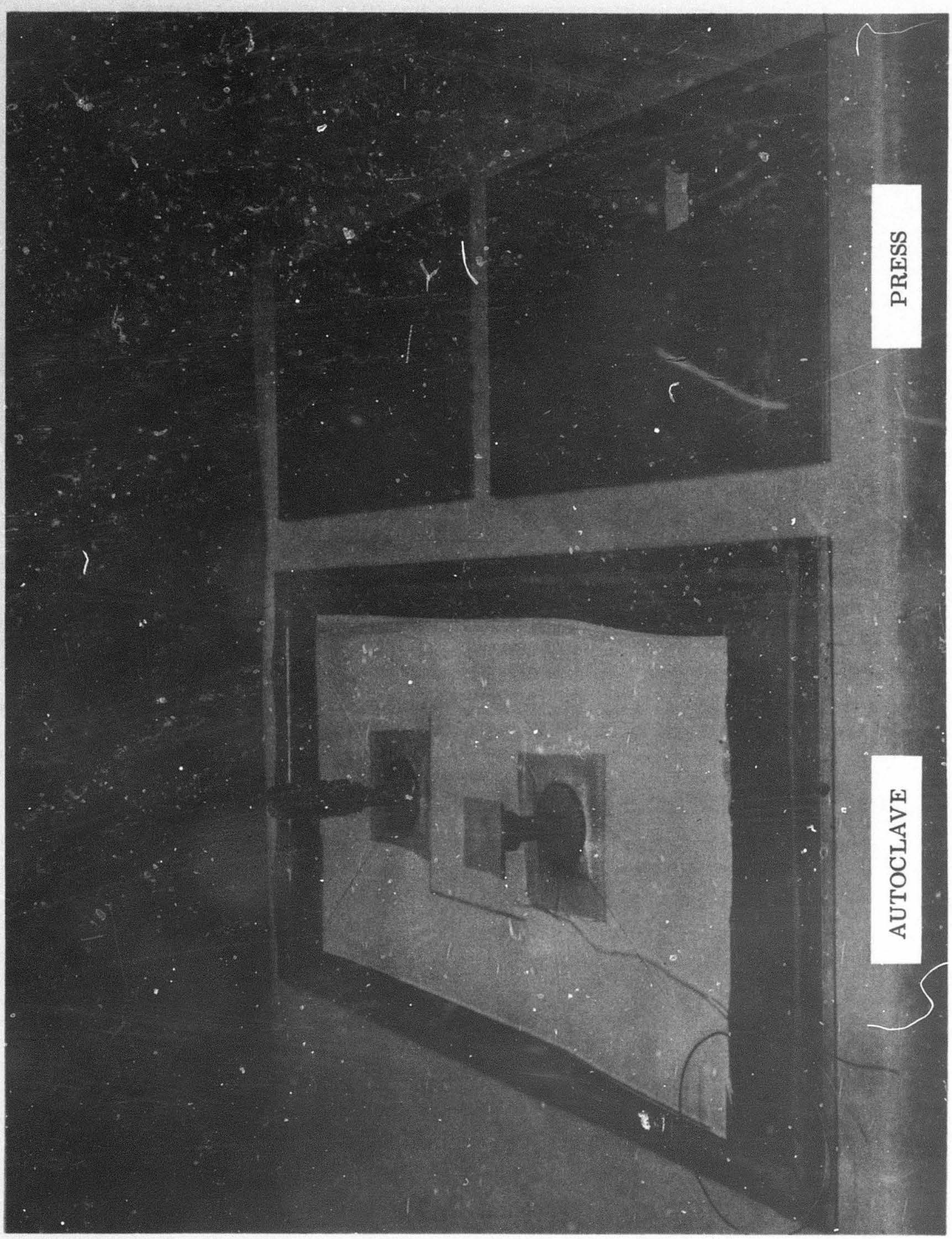



Figure 1 - Parts Prepared for Processing

Tables I, II, III, IV, V, and VI present the results of the process development study on the fabrication of four-ply polyimide face sheets by means of the hot platen press technique. The data on Tables I, II, and III are shown graphically in Figures 2, 3, and 4, respectively. Figure 5 shows the laboratory press used in this program.

An analysis of this data showed the following:

1. Application of Pressure

   The most important factor in the press fabrication of polyimide face sheets was the point at which pressure was applied during the cure cycle. As the polyimide resin underwent heat conversion, three conditions occurred: 1) the resin became quite fluid; 2) considerable outgassing occurred, and 3) gellation then took place rapidly. Pressure had to be applied to the laminate at the end of the outgassing phase after the resin began to get viscous, but before solid gellation occurred. If pressure was applied too soon, the combination of high fluidity and outgassing caused an excessive amount of resin flash. This produced a resin-starved laminate with a high void content. If the pressure was applied too late, the resin had gelled and little or no flow occurred. This produced a laminate with large internal voids, a poor surface condition, and low interlaminar adhesion.

   Figures 2, 3, and 4 show quite graphically that there was an optimum point during cure when pressure had to be applied in order to obtain the best physical properties.

2. Pressure

   Table IV shows the effect of pressure on the physical properties of press fabricated laminates. As would be expected, the properties improved as the pressure was increased. It is interesting to note, however, that - while 100 psi seemed obviously too little pressure - the difference between 250 psi and 1000 psi was not very great.

3. Temperature Profile

   Tables I, II, and III, and Figures 2, 3, and 4 show a comparison of the effect of heat-up rate on process control and reproducibility. If the heat-up rate was too rapid (Table I and Figure 2) the temperature uniformity was questionable, control was difficult and physical property results unpredictable. If the heat-up rate was slower and more uniform then the processing could be controlled and the laminate properties were reproducible (Tables II and III and Figures 3 and 4).

## TABLE I

### PRESS FABRICATION OF 4-PLY POLYIMIDE LAMINATES

### INVESTIGATION OF PRESSURE APPLICATION POINT

### INITIAL PLATEN TEMPERATURE - 600° F

| SAMPLE NO. | CONTACT TIME MIN-SECS | LAMINATE TEMP WHEN PRESSURE WAS APPLIED - ° F - | LAMINATE THICKNESS - INCHES- | RESIN CONTENT -%- | FLEXURAL STRENGTH -PSI- |
|---|---|---|---|---|---|
| 2 | -- | | .045 | 17.7 | 9,390 |
| 3 | | | .040 | 24.7 | 35,850 |
| 4 | | | .034 | 13.2 | 48,660 |
| 5 | | | .036 | 13.3 | 41,720 |
| 6 | 2-30 | | | 22.5 | 36,140 |
| 7 | 3-0 | | | 21.6 | 26,300 |
| 8 | 3-30 | | | 25.4 | 36,900 |
| 9 | 4-0 | | | 22.7 | 18,860 |
| 10 | 4-0 | | | 29.6 | 27,040 |
| 11 | 4-30 | | | 29.9 | 40,940 |
| 12 | 4-15 | | | 24.4 | 32,775 |
| 14 | 4-30 | | .042 | 20.8 | 16,560 |
| 15 | 4-15 | | .043 | 30.9 | 38,600 |
| 16 | 5-0 | | .041 | 26.0 | 30,180 |
| 27 | 0-37 | 425 | | 13.7 | 77,740 |
| 28 | 1-40 | 500 | | 24.8 | 66,600 |
| 29 | 2-45 | 575 | | 23.1 | 57,900 |
| 35 | 3-30 | 560 | | 19.5 | 47,340 |
| 36 | 3-30 | 560 | | 22.9 | 49,810 |

**NOTES:**

1. Four-ply polyimide prepreg lay-up was inserted into the press with platen preheated to 600° F.

2. Pressure used in all cases was 250 psi.

3. Control was difficult due to rapid heat-up rate of laminate.

## TABLE II

### PRESS FABRICATION OF 4-PLY POLYIMIDE LAMINATES

### INVESTIGATION OF PRESSURE APPLICATION POINT

### INITIAL PLATEN TEMPERATURE - ROOM TEMPERATURE

| SAMPLE NO. | CONTACT TIME MIN-SECS | LAMINATE TEMP WHEN PRESSURE WAS APPLIED - °F - | LAMINATE THICKNESS -INCHES- | RESIN CONTENT - % - | FLEXURAL STRENGTH -PSI- |
|---|---|---|---|---|---|
| 17 | 15-0 | 540 | .040 | 26.9 | 54,680 |
| 18 | 16-0 | 525 | .039 | 27.3 | 62,120 |
| 19 | 17-0 | 540 | .039 | 27.8 | 58,880 |
| 20 | 18-0 | 560 | .039 | 24.8 | 60,420 |
| 21 | 19-0 | 570 | .039 | 28.3 | 54,780 |
| 22 | 16-40 | 530 | .039 | 28.4 | 52,230 |
| 23 | 16-40 | 550 | .039 | 28.2 | 52,710 |
| 24 | 14-0 | 510 | .037 | 26.4 | 66,220 |
| 25 | 13-40 | 500 | .038 | 27.0 | 63,940 |
| 30 | 11-15 | 425 | -- | 27.7 | 66,200 |
| 31 | 13-0 | 500 | -- | 25.9 | 79,200 |
| 32 | 19-0 | 575 | -- | 29.2 | 55,220 |
| 37 | 25-0 | 550 | -- | 25.2 | 43,790 |
| 47A | 12-20 | 400 | .035 | 26.2 | 70,800 |
| 47B | 13-10 | 425 | .037 | 23.5 | 66,120 |
| 47C | 13-15 | 450 | .037 | 27.0 | 76,360 |
| 47D | 14-15 | 475 | 037 | 26.3 | 63,380 |
| 47E | 16-0 | 500 | .038 | 27.5 | 63,400 |
| 47F | 17-25 | 525 | .038 | 27.6 | 60,860 |

NOTES:

1. Four-ply lay-ups were placed in a room temperature press and the temperature then raised to 600° F.
2. Pressure applied in each case was 250 psi.
3. Shims were used for the 47 series. The shims were removed just prior to application of pressure.

## TABLE III

### PRESS FABRICATION OF 4-PLY POLYIMIDE LAMINATES

### INVESTIGATION OF PRESSURE APPLICATION POINT

### INITIAL PLATEN TEMPERATURE - 250° F

| SAMPLE NO. | CONTACT TIME MINS-SECS | LAMINATE TEMP WHEN PRESSURE WAS APPLIED - °F - | LAMINATE THICKNESS -INCHES- | RESIN CONTENT - % - | FLEXURAL STRENGTH -PSI- |
|---|---|---|---|---|---|
| 26 | | 575 | | 24.5 | 55,740 |
| 33 | | 500 | | 25.4 | 60,040 |
| 34 | | 425 | | 25.3 | 73,540 |
| 39 | 5-54 | 350 | .031 | 18.3 | 38,240 |
| 40 | 6-45 | 375 | .034 | 22.0 | 37,460 |
| 41 | 7-35 | 400 | .034 | 25.3 | 60,940 |
| 48A | 9-0 | 425 | .035 | 25.9 | 74,440 |
| 48B | 10-0 | 450 | .036 | 25.4 | 71,760 |
| 48C | 10-35 | 475 | .037 | 25.8 | 63,080 |
| 48D | 10-05 | 500 | .037 | 25.2 | 63,220 |
| 48E | 12-40 | 525 | .038 | 24.5 | 63,840 |

**NOTES:**

1. Pressure applied in each case was 250 psi.

2. Shims were used during the heat-up period in all cases except for samples 26, 33 and 34. The shims were removed just prior to the application of pressure.

## TABLE IV

## PRESS FABRICATION OF 4-PLY POLYIMIDE LAMINATES

### EFFECT OF PRESSURE

| SAMPLE | CONTACT TIME MINS | SECS | LAMINATE TEMP WHEN PRESS APPLIED ° F | PRESS. PSI | LAMINATE THICKNESS-INS | RESIN CONTENT % | FLEXURAL STRENGTH PSI |
|---|---|---|---|---|---|---|---|
| 49 | 7 | 30 | 425 | 100 | .038 | 27.5 | 57,450 |
| 48A | 9 | 0 | 425 | 250 | .035 | 25.9 | 74,440 |
| 50 | 8 | 20 | 425 | 1000 | .033 | 26.5 | 79,120 |

NOTES:

1. Initial platen temperature - 250° F
2. Shims were used during the heat-up period.
3. Cure temperature 600° F

## TABLE V

### EFFECT OF USING SHIMS DURING HEAT-UP AND OUTGASSING OF 4-PLY POLYIMIDE LAMINATE

| LAMINATES PREPARED WITHOUT USING SHIMS | | LAMINATES PREPARED USING SHIMS | |
|---|---|---|---|
| TEMP AT WHICH 250 PSI PRESSURE WAS APPLIED | FLEXURAL STRENGTH-PSI | TEMP AT WHICH SHIMS REMOVED & 250 PSI PRESSURE APPLIED | FLEXURAL STRENGTH |
| 425 | 66,200 | | |
| 500 | 63,940 | 400 | 70,800 |
| 510 | 66,220 | 425 | 66,120 |
| 525 | 62,120 | 450 | 76,360 |
| 530 | 52,230 | 475 | 63,380 |
| 540 | 58,880 | 500 | 63,400 |
| 550 | 52,710 | 525 | 60,860 |
| 560 | 60,420 | | |
| 575 | 55,220 | | |

NOTES:

1. In each case a 4-ply prepreg lay-up was inserted into a room temperature press and the temperature raised to 600° F.
2. 250 psi pressure was applied at the laminate temperatures indicated.
3. Note the variation in strength when no shims were used vs the readily apparent strength pattern obtained when shims were used.

## TABLE VI

## TWENTY MINUTE CURE VERSUS THIRTY MINUTE CURE

| FLEXURAL STRENGTH -PSI | |
|---|---|
| 30 MINUTE CURE | 20 MINUTE CURE |
| 66,200 | 66,120 |
| 63,940 | 63,400 |
| 62,120 | 60,860 |
| Average 64,087 | 63,460 |

NOTES:

1. Samples were prepared under same conditions except for the length of cure.
2. Each flexural strength value is the average of five test specimens

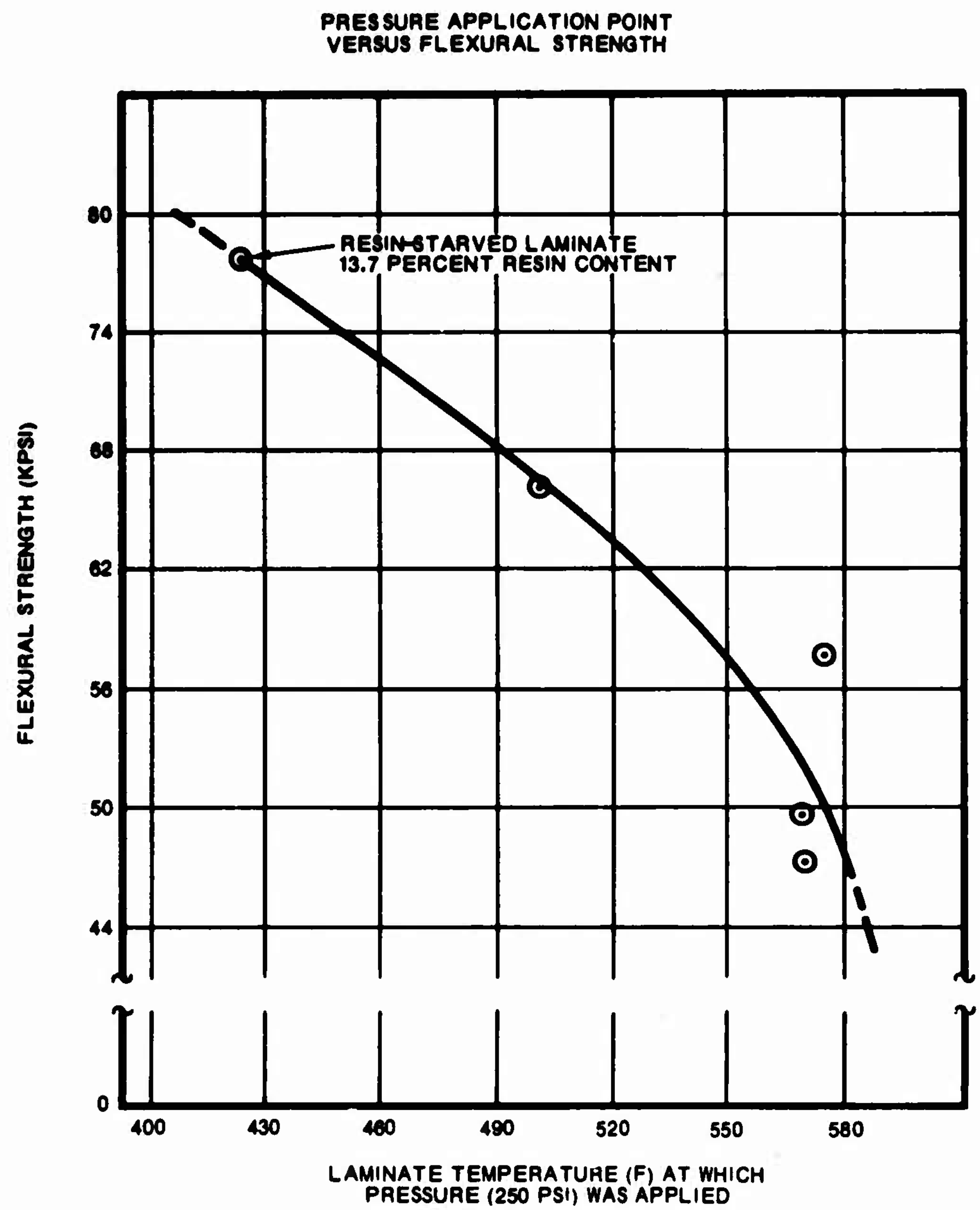



**Figure 2 - Press Fabrication of Polyimide Laminate (Initial Platen Temperature, 600 F)**

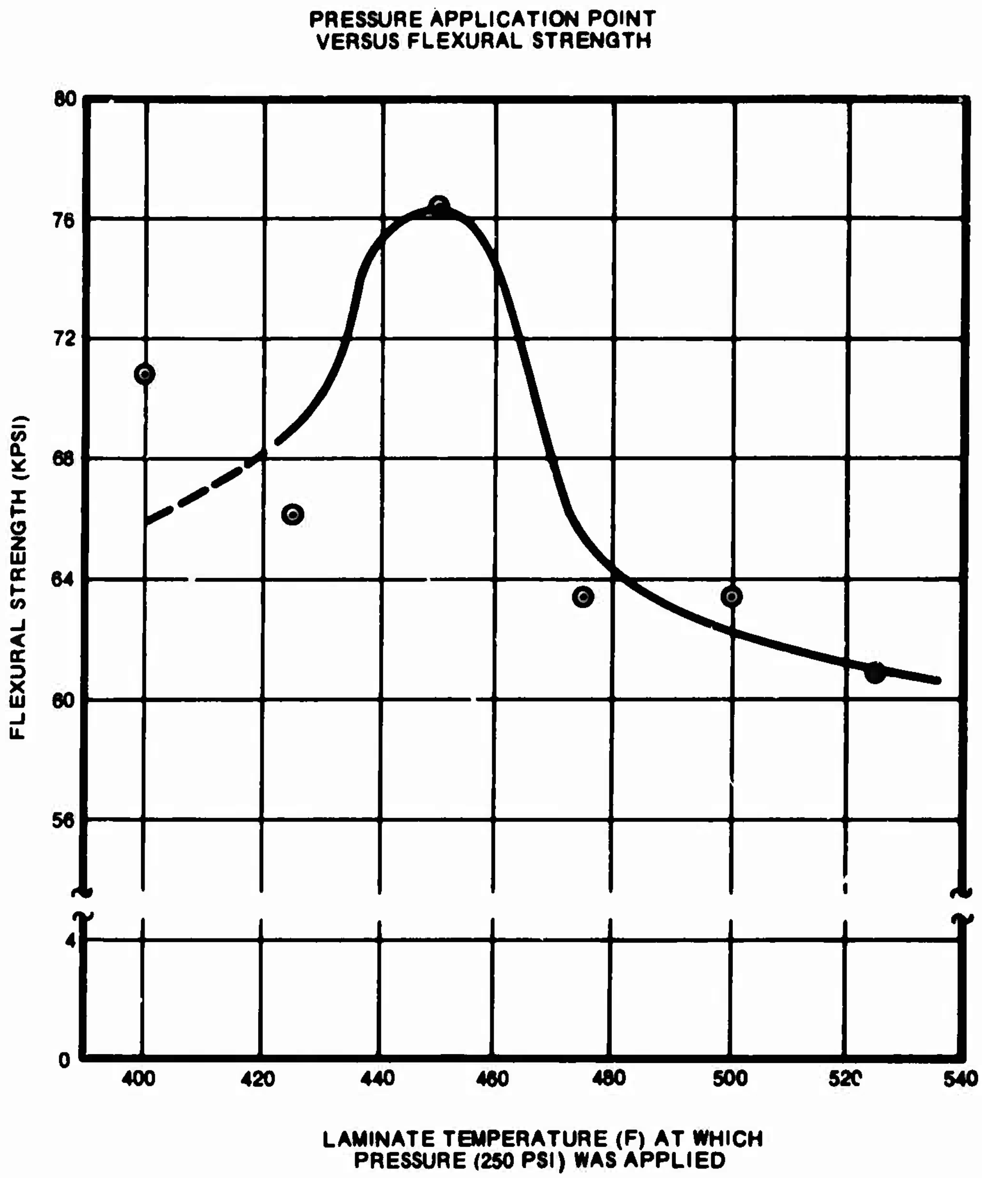



Figure 3 - Press Fabrication of Polyimide Laminate (Initial Platen Temperature, Room Temperature)

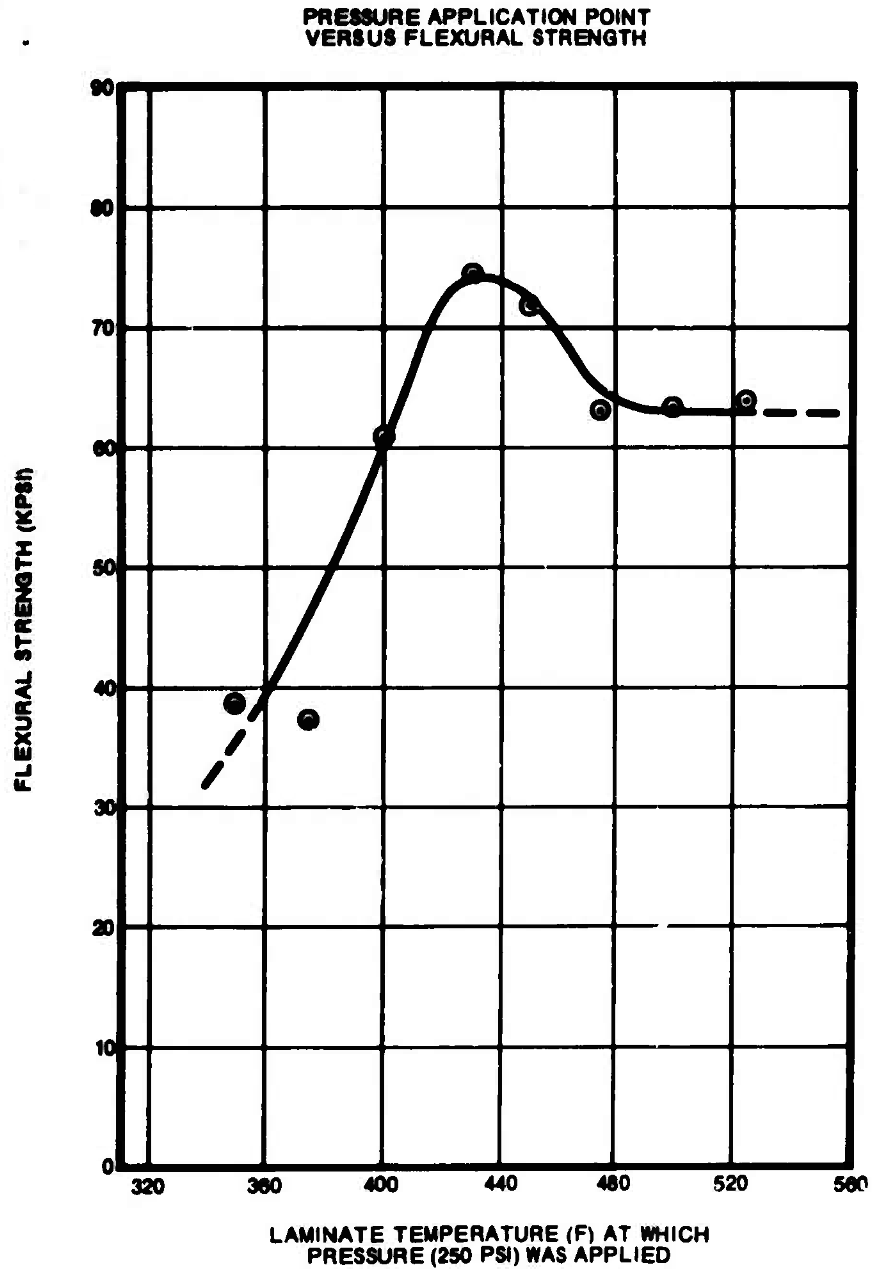



**Figure 4 - Press Fabrication of Polyimide Laminate (Initial Platen Temperature, 250 F)**

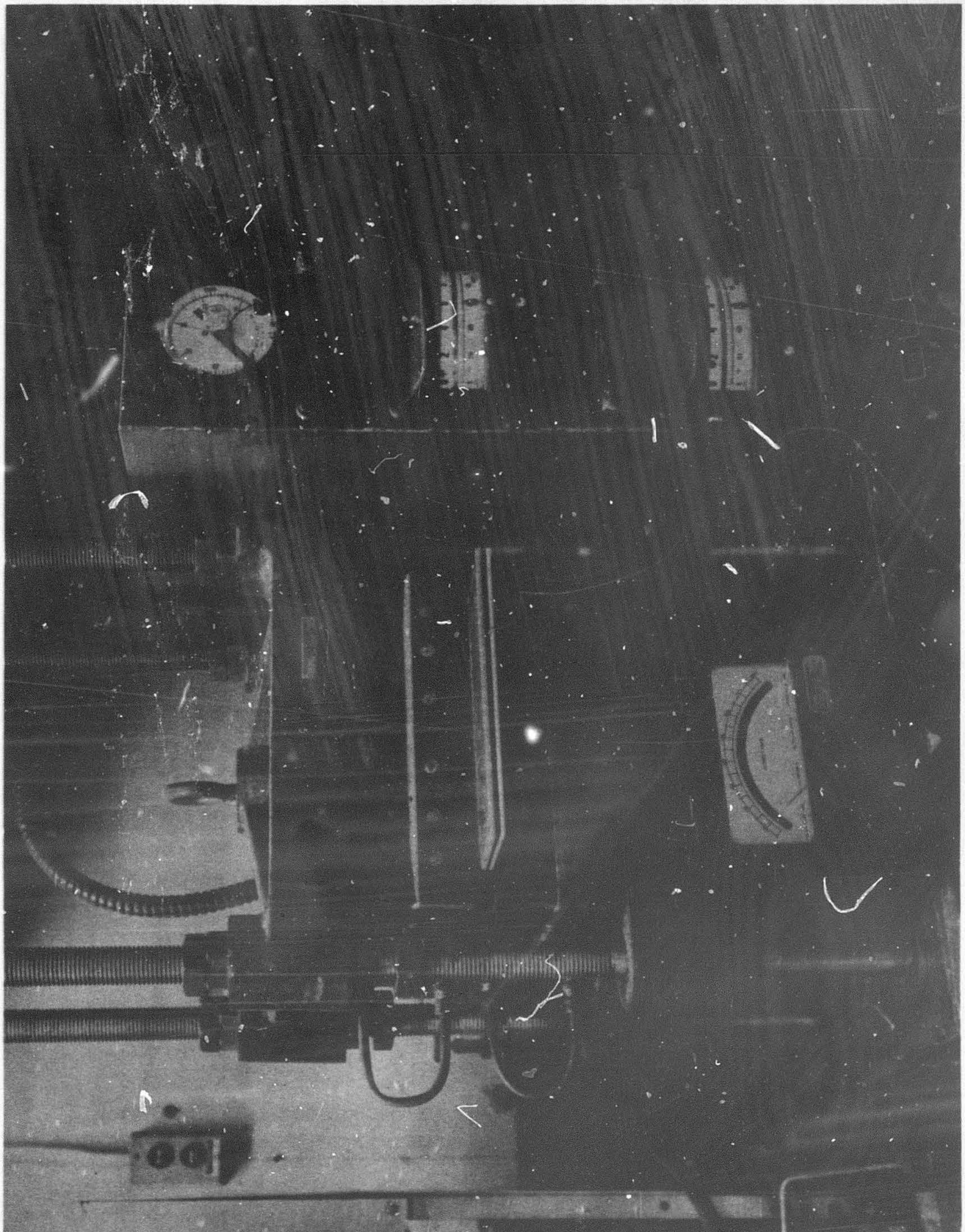


Figure 5 - Laboratory Press

4. Volatile Release

The data in Table V show that the use of shims during the heat-up and outgassing period provided an advantage over the use of unregulated "contact pressure". Unless "contact pressure" was precisely controlled, some pressure was inadvertently exerted during the outgassing phase resulting in resin blow out.

The use of shims permitted the press platens to confine the laminate sufficiently so that the heating was efficient and uniform without pressure being exerted on the plies. The spacing afforded by the shims permitted free passage of volatiles and prevented resin wash and blow out.

5. Cure Cycle

Table VI indicates that the cure time at 600F for press fabricated four-ply laminates could be reduced to 20-minutes without affecting the physical properties.

6. Recommended Press Fabricating Procedure

Based on the results of the fabricating study, the recommended processing for four-ply polyimide face sheets by the press method was established as follows:

a. Use 4-plies of 181 glass fabric reinforced polyimide prepreg.

b. Use 1-ply of 181 glass fabric release cloth on the surface eventually to be bonded to the honeycomb core.

c. Place the lay-up between aluminum caul plates treated with a RAM 225 release agent.

d. Initial press temperature should be 250F or lower.

e. Use shims that will allow the press platens to close within approximately .030" of the lay-up without exerting pressure on the lay-up.

f. Set the press temperature at 600F after the lay-up has been inserted.

g. Remove shims and pressure the lay-up to 200-250 psi at a predetermined part temperature. The temperature is read from a thermocouple in the part. The pressure point may vary depending on press heat-up rate, prepreg characteristics, and polyimide resin formulation. This pressure point must be determined for each specific set of conditions,

but once determined will remain the same as long as the conditions remain the same.

h. Cure the laminate for 20 to 30 minutes from the time of pressure application.

i. Cool under pressure to 250F.

j. No post cure is required.

k. Do not remove the release cloth peel ply until just before the face sheet is to be bonded into a sandwich composite.

## C. AUTOCLAVE FABRICATION OF POLYIMIDE FACE SHEETS

This process study included an investigation of the following factors:

1. Application of pressure.

2. Pressure.

Table VII presents the results of the process development study on the fabrication of four-ply polyimide face sheets by means of the autoclave technique. Figure 6 shows the laboratory autoclave. The knowledge concerning the behavior of the resin during cure as gained from the press fabrication study was used to simplify the autoclave study.

The heat-up was conducted at a modest and controllable rate. The laminate was covered by a perforated Teflon barrier film backed by several layers of dry bleeder cloth to provide free passage for the volatiles. Vacuum was maintained on the part throughout the heat-up and cure periods to insure smooth complete removal of the evolved gasses.

A series of experiments were performed to determine the effect of pressure and the point where pressure should be applied.

### 1. Application of Pressure

The reaction of the polyimide resin during cure in an autoclave was similar to cure in a press. The point at which pressure was applied was important. As in the case of press laminating, if the pressure was applied too late the resin did not flow properly; if the pressure was applied too soon, resin squeeze out was excessive and a resin-starved part resulted.

## TABLE VII

## AUTOCLAVE FABRICATION OF POLYIMIDE FACE SHEETS

| SAMPLE | INITIAL A/C TEMP | PRESSURE APPLICATION TIME FR START OF RUN-MINS. | PRESSURE APPLICATION LAMINATE TEMP ° F | PRESSURE APPLICATION PRESSURE PSI | RESIN CONTENT % | LAM. THICK. INCHES | FLEXURAL STRENGTH PSI |
|---|---|---|---|---|---|---|---|
| 38 | RT | | | None | 23.0 | .041 | 64,800 |
| 42 | 400 | | | None | 23.2 | .041 | 55,220 |
| 43 | 200 | At Start | 75 | 50 | 17.4 | .034 | 89,920 |
| 44 | 200 | 35' | 275 | 50 | 23.3 | .037 | 70,660 |
| 45 | 200 | 36' | 300 | 50 | 17.0 | .038 | 67,120 |
| 46 | 200 | 63' | 325 | 50 | 24.2 | .042 | 60,320 |
| 51 | 200 | 41.5' | 285 | 185 | 23.5 | .033 | 67,760 |

Figure 6 - Goodyear Aerospace Laboratory Autoclave

It was noted that often a low-resin content laminate possessed high-mechanical strength (see Sample No. 43 in Table VII). In these cases it was theorized that the pressure had caused an efficient compacting of the reinforcing fibers without damaging them and the fluidity of the resin had reduced the proportion of bonding matrix. As the proportion of matrix material was reduced, the effective shear modulus of the matrix increased allowing more load to be shifted to the reinforcement.

However, in the case of these laminates with low-resin content and high voids, the heat resistance was drastically reduced. The heat aging properties were so poor that even though the initial strengths were high, the advantage was quickly lost on long time exposure to high-temperatures. For good heat aging properties, a laminate needed to have a higher resin content and to be compact, dense, and void free.

The data obtained in this part of the program (as presented in Table VII) showed the following:

a. Acceptable 4-ply laminates could be prepared by the use of vacuum only.

b. A moderate controllable heat-up rate was necessary.

c. The application of pressure at the proper time during the cure cycle improved the laminate.

d. If the pressure was applied too soon, a resin-starved laminate resulted.

2. Pressure

Table VII showed that a pressure of 50 psi was as effective as a higher pressure of 185 psi.

Standard production autoclaves are usually limited to pressures in the range of 50 to 100 psi. The experimental autoclave (Figure 6) used for research and development programs at the Arizona Division, Goodyear Aerospace Corporation, could attain pressures as high as 550 psi. It was felt, however, that when processing procedures were carefully controlled, a pressure of 50 psi was adequate for fabricating polyimide laminates. Also, from the standpoint of establishing fabricating procedures that could be translated to production equipment, the lower autoclave pressures were more practical.

3. Recommended Autoclave Fabricating Procedure

Based on the results of the fabricating study the recommended processing for four-ply polyimide face sheets by the autoclave method was established as follows.

a. Position 4-plies of 181 glass cloth reinforced polyimide prepreg and one ply of release cloth on an aluminum base plate.

b. Cover the prepreg with a perforated teflon barrier film.

c. Cover the barrier film with several plies of bleeder cloth.

d. Use an edge bleeder around the periphery of the lay-up.

e. Position several vacuum outlets (never less than 2) on the edge bleeder.

f. Use a 5-mil nylon or Mylar bagging film.

g. Seal the film to the base plate with a high-temperature sealant.

h. Pull a vacuum on the part and seal all leaks. Leave vacuum on throughout cure cycle.

i. Insert part in the autoclave. Initial autoclave temperature should be below 250F.

j. Set autoclave temperature at 400F.

k. At a predetermined part temperature (thermocouple in part), pressure the autoclave to 50 psi. As explained in the discussion on press fabrication above, the temperature at which pressure must be applied is predetermined for each set of specific conditions which include material, molds, autoclave characteristics, etc.

l. Cure for 90 minutes from time of applying pressure.

m. Cool to below 250F under pressure and vacuum before removing from the autoclave.

n. No post cure is required for the 4-ply laminate.

o. Remove release cloth peel ply just prior to bonding into a sandwich composite.

## D. POST CURE STUDY

Data on the post cure study are presented in Table VIII. The results indicate that a post cure operation on the polyimide face sheet laminates had little effect. This was not too surprising when one considers that the face sheets were thin (.035 - .040) four-ply laminates. The resin matrix throughout the laminate was practically completely converted during the cure. It has been well established that thicker polyimide laminates require a post cure treatment to achieve optimum properties.

## E. BONDABLE SURFACE STUDY

In the secondary bond technique for making a sandwich composite, a precured face sheet had to be bonded to the core by means of an adhesive film. It was necessary to roughen the mating surface of the face sheet in order to achieve an effective bond. Several techniques were evaluated for preparing a bondable surface on a polyimide face sheet:

1. Untreated surface - Control
2. Mechanical roughening
3. Dry peel ply
4. Prepreg peel ply
5. Release cloth peel ply

Test results on the study of bondable surface preparation are presented in Table IX. The condition of the surface was evaluated by means of lap shear samples using a polyimide adhesive.

The release cloth and the dry peel ply techniques were best, as expected. A microscopic examination of the treated surfaces showed the following:

1. Control - smooth, glazed, glossy.
2. Sand blasted - rough, fibrous, loose lightly adhered particles.
3. Dry peel ply - rough, tightly adhered fibrous particles.
4. Prepreg peel ply - slightly glazed, slightly rough.
5. Release cloth - rough, peaks, and deep valleys.

TABLE VIII

POST CURE STUDY

| SAMPLE NO. | POST CURE SCHEDULE | FLEXURAL STRENGTH-PSI |
|---|---|---|
| 13-21 | Control - No Post Cure | 36,166 |
| 13-1 | a. 30 mins R.T. to 400° F | 39,266 |
| | b. 90 " at 400° F | |
| | c. 60 " " 500° F | |
| | d. 60 " " 600° F | |
| | e. 5.5 hours at 700° F | |
| 13-2 | a. 90 mins. at 400° F | 35,266 |
| | b. 60 " " 500° F | |
| | c. 60 " " 600° F | |
| | d. 5.5 hours at 700° F | |
| 13-3 | a. 60 mins. at 500° F | 34,433 |
| | b. 60 mins " 600° F | |
| | c. 5.5 hours at 700° F | |
| 13-4 | a. 60 mins. at 600° F | 40,816 |
| | b. 5.5 hours at 700° F | |
| 13-5 | a. 5.5 hours at 700° F | 34,766 |
| 13-6 | a. 30 mins. R.T. to 400° F | 38,933 |
| | b. 30 " at 400° F | |
| | c. 30 " " 500° F | |
| | d. 30 " " 600° F | |
| | e. 120 " " 700° F | |
| 13-7 | a. 30 mins. at 400° F | 37,966 |
| | b. 30 " " 500° F | |
| | c. 30 " " 600° F | |
| | d. 120 " " 700° F | |
| 13-8 | a. 30 mins. at 500° F | 37,500 |
| | b. 30 " " 600° F | |
| | c. 120 " " 700° F | |
| 13-9 | a. 30 mins. at 600° F | 37,833 |
| | b. 120 " " 700° F | |

NOTES:

1. Four-ply laminate was prepared by the autoclave technique.
2. Resin content was 24.9%
3. Thickness was .042
4. Post cures were conducted in presence of air.

## TABLE IX

### BONDABLE SURFACE STUDY

| PANEL DESIGNATION | SURFACE TREATMENT | RESIN CONTENT % | LAP SHEAR STRENGTH (PSI) |
|---|---|---|---|
| I | Control-No Preparation | 22.8 | Failed to Bond |
| II | Sand Blasted Surface | 26.8 | 701 |
| III | Dry 128 Peel Ply | 26.6 | 1109 |
| IV | 128 Prepreg Peel Ply | 26.1 | 779 |
| V | Release Cloth (1B301 F54) | 24.6 | 1844 |

**NOTES:**

1. Four-ply laminate prepared by autoclave technique.
2. Polyimide adhesive used for bonding lap shear samples.
3. Autoclave technique used to cure adhesive.
4. Lap shear strength values are average of five specimens.

The release cloth technique was used throughout the program for providing a bondable surface on polyimide face sheets used in the secondary bond fabrication method for composite structures. The release cloth was obtained from Coast Manufacturing and Supply Company under the designation "Trevarno Pink Release Cloth 1B-301-F54".

## F. COMPARATIVE STUDY OF POLYIMIDE RESINS

This study compared the performance of the following materials:

1. Monsanto Skybond 700 prepreg from Supplier A.

2. Monsanto Skybond 700 prepreg from Supplier B.

3. duPont PI-2501 prepreg (duPont - supplier).

It is pointed out that this study was based on small lot shipments of prepreg and does not necessarily represent the supplier's overall capabilities. It is generally recognized that given a standardized set of specifications and requirements, most of the commercial prepreggers can produce quality prepregs that are acceptable and comparable.

However, while this study was not intended to compare prepreg producers, it did show that the polyimide resins required skill and care in processing; that standard specifications for prepregs had not been finalized; and that prepreg characteristics, as well as process techniques, affected the laminate properties.

Table X presents the comparative data obtained from the evaluation of press fabricated laminates.

Table XI presents the comparative data obtained from the evaluation of autoclave fabricated laminates.

The method by which the laminates in this comparative study were prepared was termed the "Pressure-Point Technique". This technique proved most successful for fabricating both polyimide and polybenzimidazole laminates and sandwich composites.

In the "Pressure-Point" technique the composite was heated up at a steady controlled rate under contact pressure - using shims for a press operation or vacuum only in the case of an autoclave operation. The temperature of the composite was read accurately by means of an imbedded thermocouple

At a specified composite temperature, pressure was applied. The cure was continued under pressure and the composite was cooled under pressure. The temperature at which pressure was applied

## TABLE X

## COMPARATIVE STUDY - POLYIMIDE PREPREGS

### PRESS CURE

| SUPPLIER A (Skybond 700) | | SUPPLIER B (Skybond 700) | | PI 2501 | |
|---|---|---|---|---|---|
| Sample No. | Flex Strength | Sample | Flex Strength | Sample | Flex. Strength |
| 34 | 73,540 | 73 | 68,500 | 67 | 73,220 |
| 48A | 74,410 | 74 | 68,820 | 69 | 69,280 |
| 48B | 71,760 | 75 | 47,900 | 70 | 70,460 |
| 48C | 63,080 | 81 | 56,650 | 71 | 70,570 |
| | | 82 | 61,700 | | |
| | | 83 | 67,250 | | |

NOTES:

1. All laminates were 4-ply construction
2. Initial press temperature was 250 F.
3. Pressure was 200 psi.

TABLE XI

**COMPARATIVE STUDY - POLYIMIDE PREPREGS**

**AUTOCLAVE CURE**

| Supplier A (Skybond 700) | | Supplier B (Skybond 700) | | P.I. 2501 | |
|---|---|---|---|---|---|
| **Sample** | **Flex Strength** | **Sample** | **Flex Strength** | **Sample** | **Flex Strength** |
| 43 | 87,920 | 52 | 63,220 | | |
| 44 | 70,660 | 76 | 40,440 | 68 | 80,200 |
| 45 | 67,120 | 92 | 54,700 | 72 | 80,460 |

**NOTES:**

1. All laminates were 4-ply construction.

2. Full vacuum was applied throughout the cure.

3. Autoclave pressure used was 50 psi.

had to be determined for each set of specific conditions. This temperature depended on initial press or autoclave temperature, heat-up rate, type of resin, prepreg characteristics, etc.

In this comparative study of polyimide resins several laminates were prepared in each series in order to probe the pressure-point range and establish the optimum pressure-point temperature. Therefore, the higher values obtained were the most significant and represent optimum conditions. The low values indicated that the pressure was applied outside the optimum temperature range.

Analysis of the results of this study indicated the following:

1. All three prepregs produced acceptable laminates when fabricated by the press method.

2. "A" material and duPont PI 2501 produced laminates with high flexural strength when prepared by the autoclave method. Autoclave fabricated laminates prepared from the "B" material prepreg were approximately 20% lower in flexural strength than the other two.

3. The duPont PI 2501 prepreg appeared to have a wider optimum processing range than the other two materials.

The prepreg from Supplier A was the primary polyimide prepreg used in this study.

## SECTION IV

## PROCESS STUDY - POLYIMIDE SANDWICH COMPOSITES

### A. GENERAL

The techniques for fabricating sandwich composites studied in this program were:

1. Secondary Bond - In this method the face sheets were precured and then bonded to the core by means of an adhesive layer in a secondary operation. Both the press method and the autoclave method were evaluated for producing the secondary bond.

2. Single Stage Operation - In this method both face sheets were cured and bonded to the core in a single operation. A formulated adhesive film was incorporated between the face sheet plies and the core to provide a core-to-facing bond. Some sandwich constructions were made without the adhesive film in order to evaluate the face sheet resin as a core-to-facing adhesive. Because of the high resin flow, the need for controlled uniform pressure, and problem of volatile removal only the autoclave method was used for the study of the single stage operation.

3. Multiple Stage Operation - In the study, also, only the autoclave method was employed. In this operation the outer face sheet was fabricated, then the core and inner face sheet added in one or two additional steps.

The honeycomb core stock used in this program was manufactured by Hexcel Products, Inc. The core resin system was Skybond 700 and the reinforcement was E-glass fabric. Two core densities - four pounds per cubic foot and 8 pounds per cubic foot - were evaluated. In both cores the cell size was 3/16-inch and the thickness was 1/2-inch. The cores were procured under the following designations:

"HRH-324 (Skybond 700 Resin) 3/16" cell 4.0 lb. density .500" thick."

"HRH-324 (Skybond 700 Resin) 3/16" cell 8.0 lb. density .500" thick."

The adhesive used for the polyimide sandwich composites was manufactured by the Bloomingdale Dept., American Cyanamid Company. The adhesive was composed of a modified Skybond 700 polyimide resin on a fiberglass cloth carrier. It was procured with the designation:

"FM-34 Polyimide Film Adhesive .135 lb/ft.$^2$."

## B. SANDWICH PANEL FIXTURE

Because of the high cost of the materials used in this program - particularly the honeycomb core stock - it was imperative that efficient use be made of materials. A fixture, Figure 7, was designed for the fabrication of sample sandwich composites. The fixture was designed to simulate good plant practice for fabricating sandwich structures and, at the same time, provide a sample panel from which the maximum number of test specimens could be cut with a minimum of waste. Figures 8, 9, and 10 show the fixture during various stages of laying-up a composite for single stage cure.

Figure 11 presents a sketch of the fixture in use.

The frame was designed to simulate the beveled edge of a honeycomb sandwich panel. A 6" × 12" × 1/2" sandwich composite was laid-up inside the frame. A perforated Teflon barrier film was laid over the composite and frame. Dry bleeder cloth was laid over the entire assembly and beyond the edge of the frame to form a periphery bleeder. Two vacuum connectors were situated on the periphery bleeder at the ends of the frame. The nylon or aluminum vacuum bag was sealed to the base plate by means of a high-temperature sealant.

The assembly simulated good plant practice for fabricating honeycomb core sandwich parts. The 6" × 12" × 1/2" sample panel was 100% usable for preparation of test specimens. There was no waste of materials. This fixture was used so successfully in the Phase I processing study on sandwich composites that similar fixtures were made and used for preparing the heat aging specimens for Phase II of this program.

## C. SECONDARY BOND TECHNIQUE

The process study on polyimide face sheets had shown that acceptable face sheet laminates could be prepared by both the press method and the autoclave method. Therefore, face sheets were prepared by both methods for the study of the secondary bond technique. The optimum fabricating processes as developed in the processing studies were employed.

Table XII presents data on polyimide laminates fabricated by the press method for use as face sheets on secondary bonded sandwich panels. The results emphasize the necessity for accurate and uniform process equipment and for precise control of process conditions. These laminates were larger than the ones prepared during the processing studies and nearly covered the entire press platen area. This altered the heating pattern and the thermocouple location from that used during the early processing studies. It was necessary to re-establish the optimum temperature range. Figure 12, which is a graph of the values in

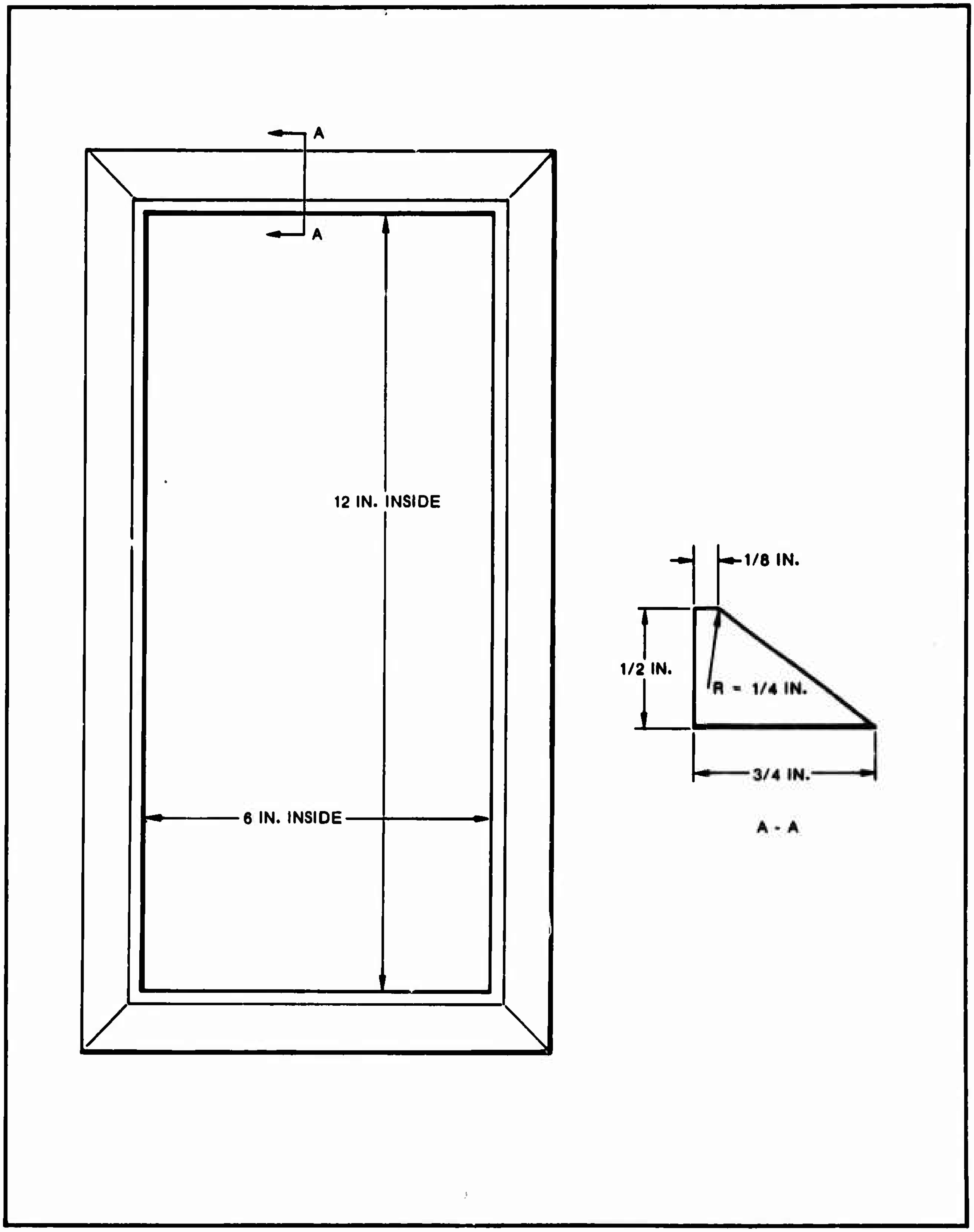



Figure 7 - Sandwich Panel Fixture

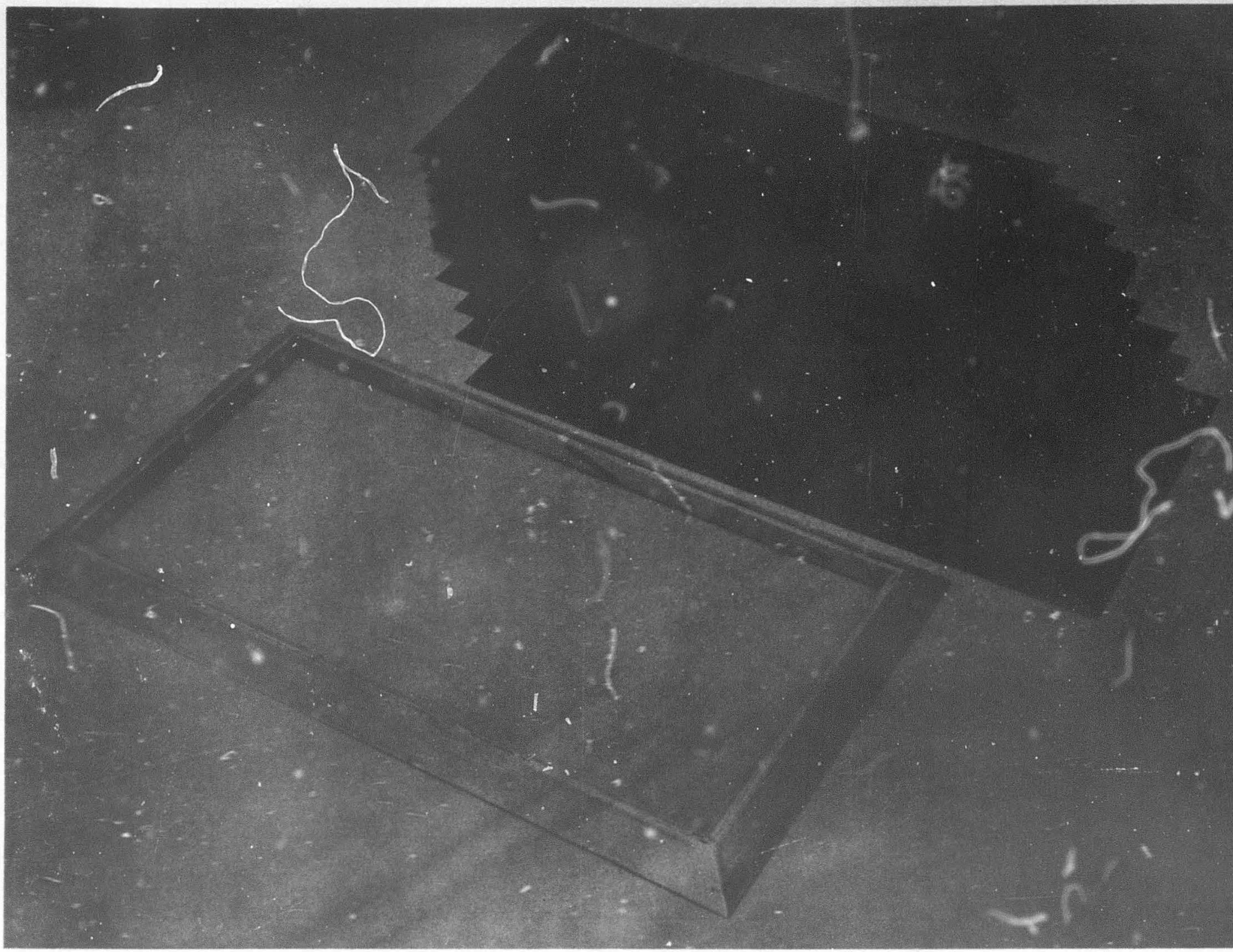


Figure 8 - Sample Sandwich Composite Fixture

Figure 9 - Sandwich Fixture During Layup

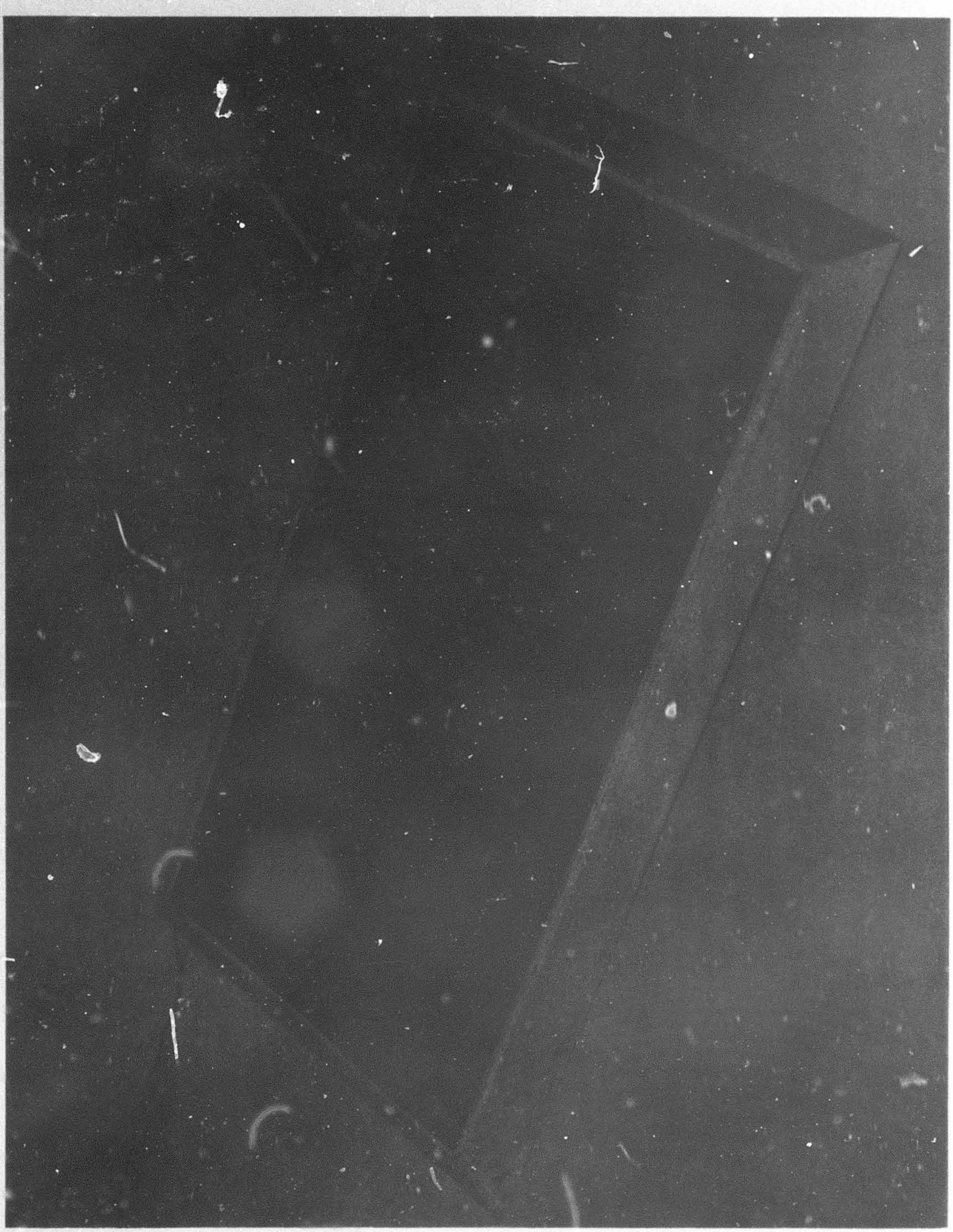


Figure 10 - Sandwich Fixture Ready for Bagging

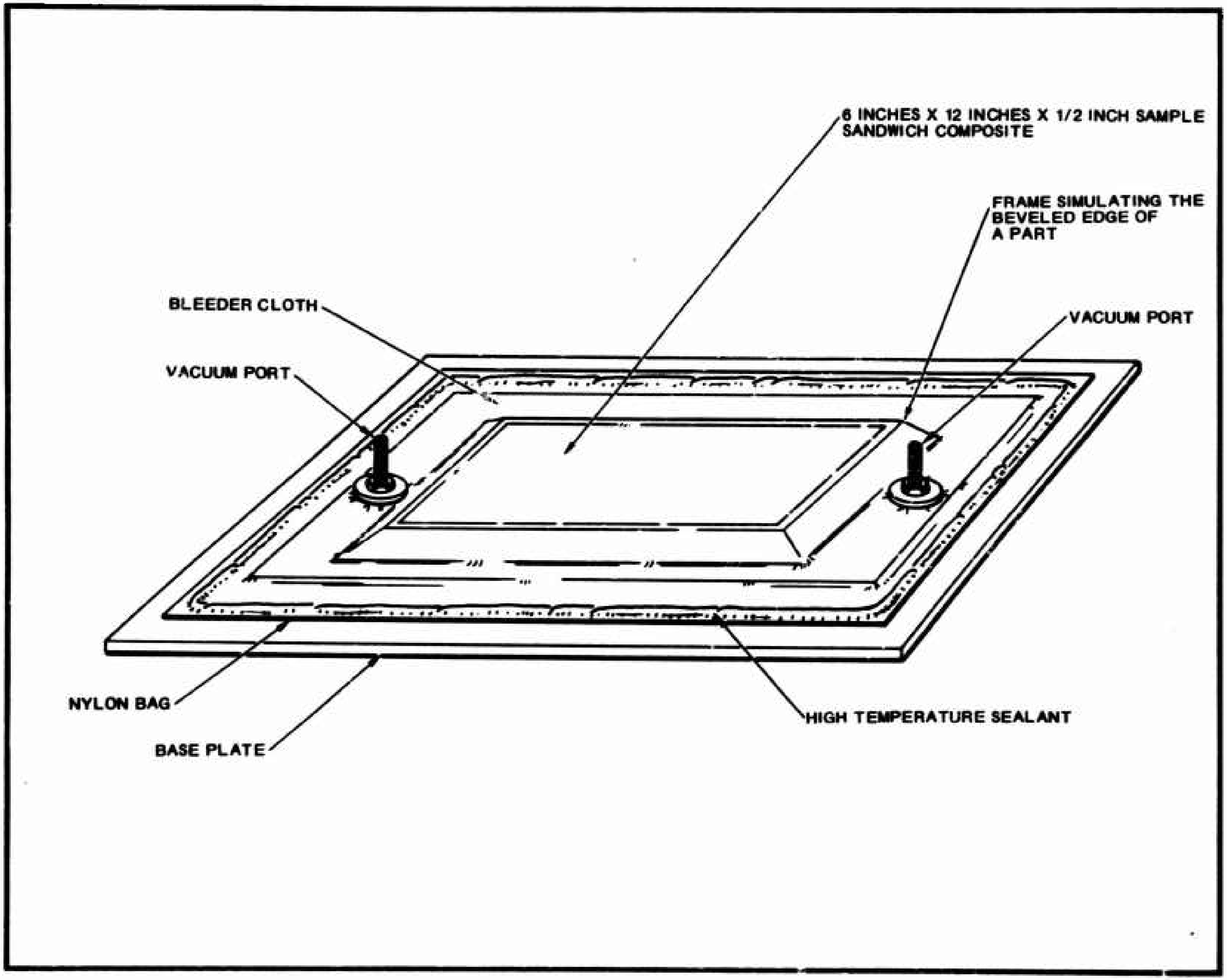



Figure 11 - Assembly for Preparing Sandwich Composites

## TABLE XII

**PRESS FABRICATION OF POLYIMIDE FACE SHEETS FOR SECONDARY BONDING**

| SAMPLE NO. | LAMINATE TEMP. WHEN PRESS. WAS APPLIED °F | THICKNESS INCHES | R.C. % | FLEXURAL STRENGTH PSI |
|---|---|---|---|---|
| 84 | 425 | .041 | 26.4 | 53,550 |
| 85 | 425 | .040 | 25.4 | 57,750 |
| 86 | 400 | .034 | 22.5 | 79,820 |
| 99 | 425 | .042 | 25.7 | 43,960 |
| 100 | 400 | .039 | 23.5 | 69,600 |
| 102 | 380 | .034 | 23.5 | 77,625 |
| 103 | 410 | .041 | 26.2 | 54,700 |

**NOTES:**

1. Initial platen temperature was 250 F.
2. Pressure applied was 200 psi.
3. All laminates were 4-plies.
4. All laminates had a release cloth peel ply on one surface.

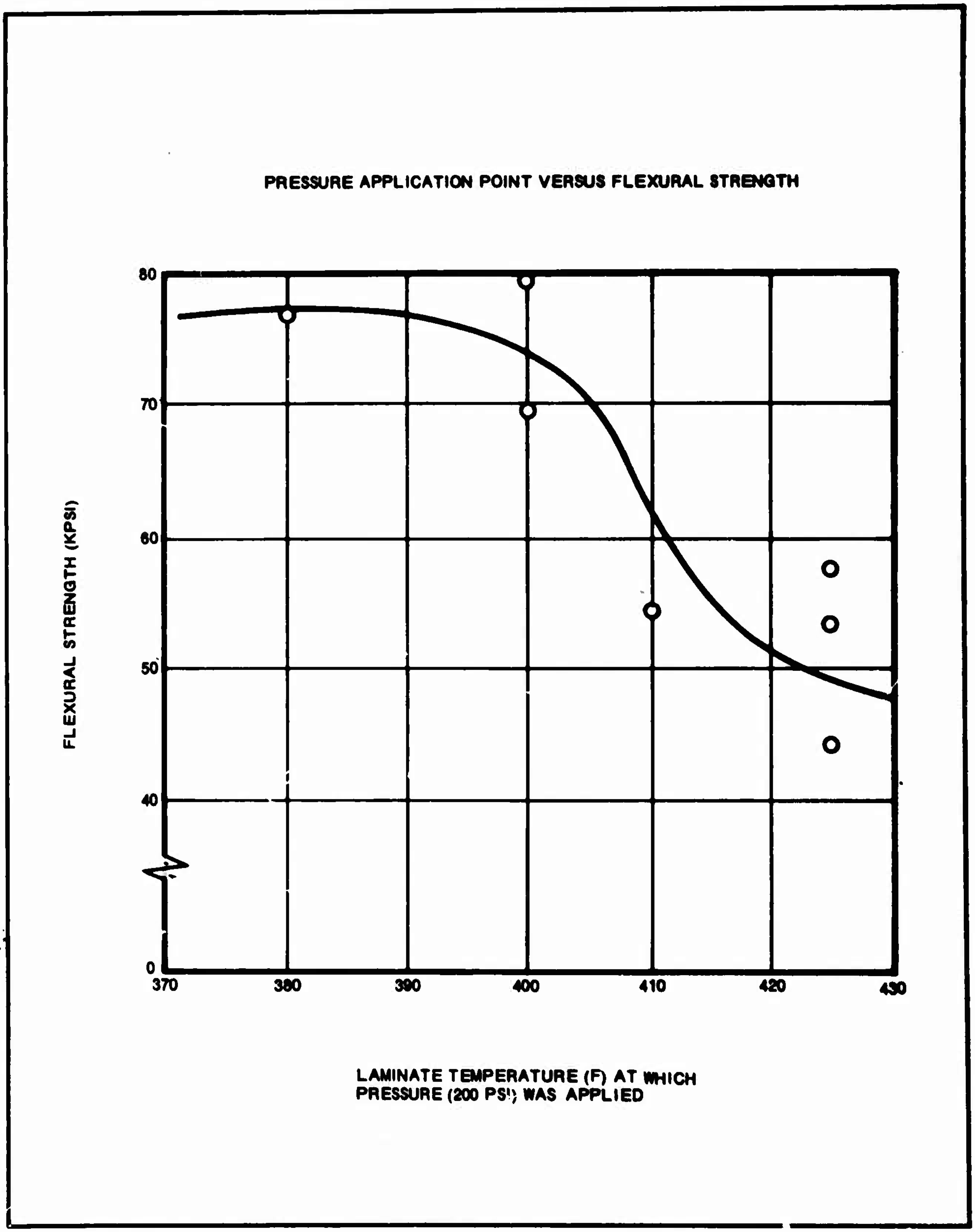



**Figure 12 - Press Fabrication of Polyimide Face Sheets for Secondary Bonding**

Table XII, shows that an optimum pressure-point range existed but was shifted to a lower temperature than previously established. This illustrated the fact that the pressure-point range was influenced by many processing factors and had to be established for each set of process conditions.

Table XIII presents data on polyimide face sheets fabricated by the autoclave method.

The secondary bond technique for preparing sandwich composites was performed in the following manner:

> Four-ply laminates prepared by either the press or autoclave method were used. A ply of release cloth had been included in the lay-up and was laminated to one surface of each skin to serve as a peel ply. The peel ply was removed from the cured laminates, exposing a bondable surface. These skins were bonded to the honeycomb core material by means of a layer of polyimide film adhesive. Both skins were bonded to the core at the same time. Either a press method or an autoclave method was used to supply the heat and pressure required for the bonding operation.

Table XIV presents the results of the study on polyimide sandwich composites prepared by means of the secondary bond technique.

## D. SINGLE STAGE TECHNIQUE

In this technique, all parts of the sandwich composite were laid-up and cured in an autoclave at the same time. Four plies of polyimide prepreg were laid-up for the skins. No peel ply was used in this technique. A layer of film adhesive was laid between the skin prepreg plies and the core. (Note: In several of the composites the film adhesive was omitted in order to determine if the face sheet resin could serve as the core to facing adhesive.) The entire assembly was covered with a perforated barrier film and bleeder cloth and sealed in a vacuum bag fitted with vacuum outlets. The assembly was placed in an autoclave. Vacuum was maintained on the part throughout the cure. Pressure was applied to the autoclave at a predetermined part temperature (pressure-point cure technique).

Table XV presents data generated during the study on the fabrication of polyimide sandwich composites by means of the single stage technique. Based on the results of the study, the recommended processing for a single stage cure polyimide honeycomb core sandwich composite was determined to be as follows:

1. Position 4-plies of 181 glass cloth reinforced polyimide prepreg (lower face sheet lay-up) on an aluminum base plate.

## TABLE XIII

**AUTOCLAVE FABRICATION OF POLYIMIDE FACE SHEETS FOR SECONDARY BONDING**

| SAMPLE | LAMINATE TEMP. WHEN PRESS. WAS APPLIED °F | THICKNESS INCHES | PRESSURE PSI | R.C. % | FLEX PSI |
|---|---|---|---|---|---|
| 101 | 270 | .035 | 50 | 20.8 | 66,480 |
| 104 | 275 | .033 | 50 | 21.2 | 83,360 |
| 116 | 275 | .033 | 50 | 20.6 | 52,940 |
| 117 | 280 | .032 | 100 | 19.1 | 49,150 |
| 118 | 280 | .033 | 100 | 19.0 | 57,200 |

NOTES:

1. Laminates were 4-ply construction.
2. Full vacuum applied throughout cure.
3. All laminates had a release cloth peel ply on one surface

TABLE XIV

POLYIMIDE SANDWICH COMPOSITES - SECONDARY BOND

| Sample | Prepreg Sup-plier | Adhe-sives | Method of Prep. Skins | Method of Prep. Sandwich | Flatwise Tensile | Core Shear | Flex Force lb/in Width |
|---|---|---|---|---|---|---|---|
| 3 | B | FM34 | Press | Press | 278 | 242 | 262 |
| 5 | B | FM34 | Press | A/C | 242 | 227 | 245 |
| 7 | A | FM34 | Press | Press | 259 | 178 | 192 |
| 8 | A | FM34 | A/C | Press | 349 | 212 | 229 |
| 9 | A | FM34 | A/C | A/C | 179 | 169 | 183 |
| 14 | A | FM34 | A/C | Press | 428 | 221 | 238 |

## TABLE XV

### POLYIMIDE SANDWICH COMPOSITES - SINGLE STAGE

| Sample | Supplier Prepreg | Adhesive | Flatwise Tensile | Core Shear | lb/in Width |
|---|---|---|---|---|---|
| 2 | B | None | 289 | 301 | 316.5 |
| 4 | B | FM34 | 572 | 445 | 472 |
| 6 | C | FM34 | 653 | 263 | 280 |
| 10 | A | FM34 | 625 | 332 | 352 |
| 11 | C | FM34 | 526 | 298 | 319 |
| 12 | A | None | 224 | 226 | 240 |
| 13 | C | None | 273 | 234 | 248 |
| 15 | C | PI2501 | 339 | 270 | 281 |

NOTES:

1. Optimum processing conditions were used.
2. Skins were four-ply construction.
3. Honeycomb core - HRH-324, 4.0 lbs. density, 0.50" thick.

2. Place one-ply of polyimide film adhesive on the prepreg.

3. Position the polyimide honeycomb core on the adhesive film. The core should be cleaned in a vapor degreaser to remove any possible contamination.

4. Place a ply of polyimide film adhesive on the core.

5. Position 4-plies of polyimide prepreg (upper face sheet lay-up) on the adhesive film.

6. Cover the entire composite with a perforated teflon barrier film.

7. Cover the barrier film with several plies of bleeder cloth.

8. Use an edge bleeder around the periphery of the lay-up.

9. Position several vacuum outlets (never less than 2) on the edge bleeder.

10. Use a 5 mil nylon or Mylar bagging film.

11. Seal the film to the base plate with a high-temperature sealant.

12. Pull a vacuum on the part and seal all leaks. Leave vacuum on throughout cure cycle.

13. Insert part in the autoclave. Initial autoclave temperature should be below 250F.

14. Set autoclave temperature at 400F.

15. At a predetermined part temperature (thermocouple in part) pressure the autoclave to 50 psi. ("Pressure-Point technique").

16. Cure for 90 minutes from time of applying pressure.

17. Cool to below 250F under pressure and vacuum before removing from the autoclave.

18. Post-cure the entire composite according to the following schedule:

Preheat oven to 350 F
12 minutes at 350F
12 minutes at 390 F
12 minutes at 430 F
12 minutes at 470 F
12 minutes at 510 F
2 hours at 550 F

## E. MULTIPLE STAGE TECHNIQUE

Two different multiple-stage techniques were evaluated in this study.

1. Two-Stage Method - One skin was laid-up (with release cloth peel ply) and cured in the autoclave in the normal manner for skin laminates. The peel ply was removed from the cured skin laminate and the remainder of the sandwich components were laid-up on the prepared skin; adhesive layer, honeycomb core, adhesive layer, four-plies of skin prepreg. This assembly was covered in the normal manner with a barrier film, bleeder cloth, and sealed vacuum bag. Cure was performed in the autoclave using the pressure-point technique.

   The two-stage method was investigated to provide information that could eventually prove valuable for production planning. Some aerospace plastic sandwich components require a surface skin that is aerodynamically smooth. The purpose for the two-stage technique is to obtain one skin of the sandwich composite (in most cases, the outer surface skin) as dense and void-free as possible. In the two-stage system the outer skin can be laminated at pressures well above the crushing point of the honeycomb core.

2. Three-Stage Method - In this technique, the outer skin was laid-up in the initial stage in the same manner as described previously for the two-stage method. In the second stage, the core was bonded to the bondable surface of the outer skin by means of a film adhesive.

   In the third stage, a layer of adhesive was laid over the bonded core, skin prepreg plies were added, the assembly was covered with barrier film, bleeder cloth and sealed bag, and cure performed in an autoclave using the pressure-point technique.

   The three-stage technique is also production oriented. In many cases, the shape of an aerospace vehicle component is such that the sandwich composite cannot be assembled at one time. The skin prepreg plies must be cut and fit; the core must be cut, preformed, and carefully tacked in place; edge

bands and doublers must be assembled and positioned. The three-stage system was evaluated in this program to determine feasibility and effect on properties of the composite.

Table XVI presents a comparison of the physical properties of sandwich composites prepared by secondary bond, single stage, two-stage, and three-stage.

## F. ANALYSIS OF THE POLYIMIDE SANDWICH FABRICATION STUDY.

Analysis of the data in Tables XIV, XV, and XVI indicated the following:

1. The single-stage method produced the best polyimide sandwich composites. The appearance, as well as the physical properties of the composites, was notably improved when the skin laminates and the adhesive bonds were formed simultaneously in a single operation.

   It was decided to use the single-stage method to form the polyimide sandwich composites that were subjected to the heat aging studies in Phase II of this program.

2. In the single-stage method, a prepared adhesive film was required for optimum bond between the skins and the core. An adhesive film, with controlled flow and wetting characteristics built in, provided better filleting and a better bond to the core than when the resin in the prepreg skins was used as the adhesive.

   The polyimide sandwich composites prepared for the Phase II study used the FM-34 adhesive film.

3. The secondary bond technique and the multiple stage techniques, while not as good as the single-stage method, were feasible and did produce acceptable polyimide sandwich composites.

4. In the secondary bond method, skins prepared by the autoclave technique appeared to have slightly better adhesion to the core than skins prepared by the press method. The flow of volatiles and resin through the release cloth and bleeder plies may have contributed to this factor. Peeling the release cloth from the laminate produced a rugged fibrous bonding surface.

5. Secondary bonds prepared in the press had slightly better physical properties than secondary bonds prepared in the autoclave. This was probably due to the higher temperatures associated with the press operation.

TABLE XVI

POLYIMIDE SANDWICH COMPOSITES - COMPARISON OF AUTOCLAVE TECHNIQUES

| Sample | Processing | Flatwise Tensile PSI | Core Shear PSI |
|---|---|---|---|
| 8 | Secondary Bond | 349 | 212 |
| 10 | Single Stage | 625 | 332 |
| 16 | Two-Stage | 416 | 235 |
| 17 | Three-Stage | 347 | 246 |

# SECTION V

## PROCESS STUDY - POLYBENZIMIDAZOLE FACE SHEETS

### A. GENERAL

The resin system specified for this program was AFR-151 (PBI). However, prior to the start of this contract a new PBI resin - designated PNABI - was announced. Initial physical and heat-aging properties of the new resin were quite exciting and an effort was made to include PNABI resin in this program as a substitute for the originally specified AFR-151 (PBI) resin.

Contact with the PNABI resin supplier, Narmco R and D Division, Whittaker Corp., established the fact that the new resin could not be supplied in sufficient quantity soon enough to be included in the current program. The decision was made, therefore, to continue with the AFR-151 (PBI) resin.

The AFR-151 (PBI) resin system was obtained in prepreg form on 181 E-glass reinforcement from the Narmco Materials Division, Whittaker Corp. The material designation was "PBI 2803-1581 E-glass". The investigation of the PNABI resin caused a delay in delivery of the AFR-151 (2803) PBI materials. Prior to receipt of the AFR-151 prepreg, therefore, some work was performed with the original commercial PBI prepreg, Imidite 1850 (2801-AFR-100 resin). This material was readily available and it was felt that general handling and processing information could be obtained from the 2801 prepreg since the two AFR resins (AFR-100 and AFR-151) were similar in reaction characteristics.

### B. PRELIMINARY STUDY - AFR-100 RESIN

As mentioned in the previous section, while waiting for the delivery of the AFR-151 (2803) prepreg, it was felt that some general processing information could be obtained by working with Imidite 1850 (2801 prepreg, AFR-100 resin).

The technical literature on the PBI resins generally recommended a step-cure technique. In this technique the temperature of the lay-up was raised in stages and held at a predetermined temperature for an extended period of time before raising to the next temperature plateau. In the press cure evaluation of Imidite 1850, both the step-cure and the pressure-point cure methods were investigated.

Table XVII presents the data collected during the process development study of the step-cure method as recommended by the literature.

## TABLE XVII

## PRESS FABRICATION OF 4-PLY AFR-100 (PBI) LAMINATES

## STEP-WISE CURE

| SAMPLE | CURE CYCLE Time Mins. | Press Temp | Lamin Temp | Total Dwell, Time-Mins. | Laminate Thick. Ins | Resin Content % | Flexural Strength-PSI |
|---|---|---|---|---|---|---|---|
| 53 | 30 | 350 | 341 | | | | |
| | 30 | 400 | 387 | | | | |
| | 20 | 450 | 433 | | | | |
| | Pressured to 200 psi | | | 80 | | | |
| | 20 | 500 | 475 | | | | |
| | 15 | 550 | | | | | |
| | 15 | 600 | | | | | |
| | 10 | 650 | | | | | |
| | 180 | 700 | 685 | | .052 | 39.9 | 71,270 |
| 54 | 9 | 350 to 520 | 450 | | | | |
| | 30 | 520 | 501 | | | | |
| | Pressured to 200 psi | | | 39 | | | |
| | 180 | 700 | 680 | | .047 | 32.2 | 94,960 |
| 61 | 10 | 350 to 565 | 500 | | | | |
| | 15 | 565 | 545 | | | | |
| | Pressured to 200 psi | | | 25 | | | |
| | 180 | 700 | - | | .043 | 35.9 | 89,800 |
| 62 | 8 | 350 to 540 | 450 | | | | |
| | 4 | 540 | 500 | | | | |
| | 5 | 500 | 500 | | | | |
| | Pressured to 200 psi | | | 17 | | | |
| | 180 | 700 | 690 | | .042 | 36.4 | 64,460 |
| 66 | 13 | 350 to 510 | 490 | | | | |
| | 5 | 510 | 495 | | | | |
| | Pressured to 200 psi | | | 18 | | | |
| | 180 | 700 | 700 | | .044 | 36.6 | 59,340 |

Table XVIII presents data of the study on pressure application points as developed for the polyimide resin and discussed in Section III-F of this report. The PBI resin reacted during cure in a manner similar to the polyimide resin. The cure temperature for PBI and the laminate temperature at the optimum point for application of pressure was much higher than for polyimide. Therefore the initial press temperature could be maintained at a higher temperature, 350F.

Table XIX presents a resume of the results of the process study on PBI 2801 using the press fabrication method. Figure 13, which is based on values taken from Table XIX showed that the pressure-point technique was valid and applicable to the press fabrication of PBI 2801. There was an optimum point during cure when pressure should be applied to obtain the best physical properties.

The data indicate that the more exacting stepwise cure technique did not produce a superior laminate over the controlled pressure-point technique in the case of four-ply laminates. The pressure-point method was therefore used in this program.

A comparison of Figure 13 with Figure 4 shows the similarity in cure behavior between PBI and Polyimide.

Table XX presents the results of the autoclave processing study on PBI 2801. The results showed that pressure was essential to the preparation of acceptable PBI (2801) laminates.

One of the serious problems encountered in the high-temperature autoclave processing of PBI was failure of the vacuum bag enclosing the part. Sample 56 showed that it was absolutely necessary to eliminate any air leaks in the vacuum bag during processing of PBI laminates. This was demonstrated and substantiated a number of times in processing studies on PBI. Any leaks in the vacuum bag caused excessive oxidation of the resin producing a laminate that was dark and discolored, porous, weak, with low interlaminar adhesion.

Much of the initial effort with PBI was in attempting to develop a suitable bagging technique. It was desired to have a bag that was low cost, simple to prepare and would successfully withstand the high-temperatures involved. Various materials were evaluated as high-temperature bags. The most promising proved to be a soft aluminum foil.

A secondary problem was in the use of a sealant, or method of sealing the bag, that would withstand the high-temperatures reached during the cure. Silicone sealants proved to be a satisfactory solution to this problem.

TABLE XVIII

PRESS FABRICATION OF 4-PLY AFR-100(PBI) LAMINATES

PRESSURE APPLICATION POINT STUDY

| SAMPLE | CONTACT TIME MIN -SEC. | LAM. TEMP AT TIME OF PRESSURE ° F | LAMINATE THICKNESS IN. | RESIN CONTENT % | FLEXURAL STRENGTH PSI |
|---|---|---|---|---|---|
| 55 | 10 - 25 | 550 | .034 | 27.8 | 63,375 |
| 60 | -- | 600 | .044 | 36.8 | 84,120 |
| 58 | 17 - 15 | 615 | .044 | 37.5 | 90,440 |
| 65 | 11 - 20 | 625 | .045 | 36.1 | 88,640 |
| 64 | 12 - 15 | 640 | .043 | 35.8 | 87,200 |
| 57 | 19 - 0 | 660 | .046 | 36.2 | 69,440 |

NOTES:

1. Initial platen temperature 350° F
2. Platens raised to 700° F after insertion of the part.
3. Shims used during heat-up period.
4. Cure: 90 minutes after application of pressure.

## TABLE XIX

**PRESS FABRICATION OF AFR-100 (2801 or Imidite 1850) PBI LAMINATES**

| SAMPLE | TYPE CURE | THICKNESS INCHES | R.C. % | STRENGTH FLEXURAL PSI |
|---|---|---|---|---|
| 53 | Step-Cure | .052 | 39.9 | 71,270 |
| 54 | Step-Cure | .047 | 32.2 | 94,960 |
| 61 | Step-Cure | .043 | 35.9 | 89,800 |
| 62 | Step-Cure | .042 | 36.4 | 64,460 |
| 66 | Step-Cure | .044 | 36.6 | 59,340 |
| 55 | Pressure Point (550F) | .034 | 27.8 | 63,375 |
| 57 | Pressure Point (680F) | .046 | 36.2 | 69,440 |
| 58 | Pressure Point (615F) | .044 | 37.5 | 90,440 |
| 60 | Pressure Point (600F) | .044 | 36.8 | 84,120 |
| 64 | Pressure Point (640F) | .043 | 35.8 | 87,200 |
| 65 | Pressure Point (625F) | .045 | 36.1 | 88,640 |
| 79 | Pressure Point (500F) | .044 | - | NG(Starved) |

**NOTES:**

1. All laminates were 4-ply construction.
2. The initial press temperature was 350F.
3. The temperatures recorded under column heading "Type Cure" indicate the laminate temperature when pressure was applied.
4. The pressure was 200 psi.

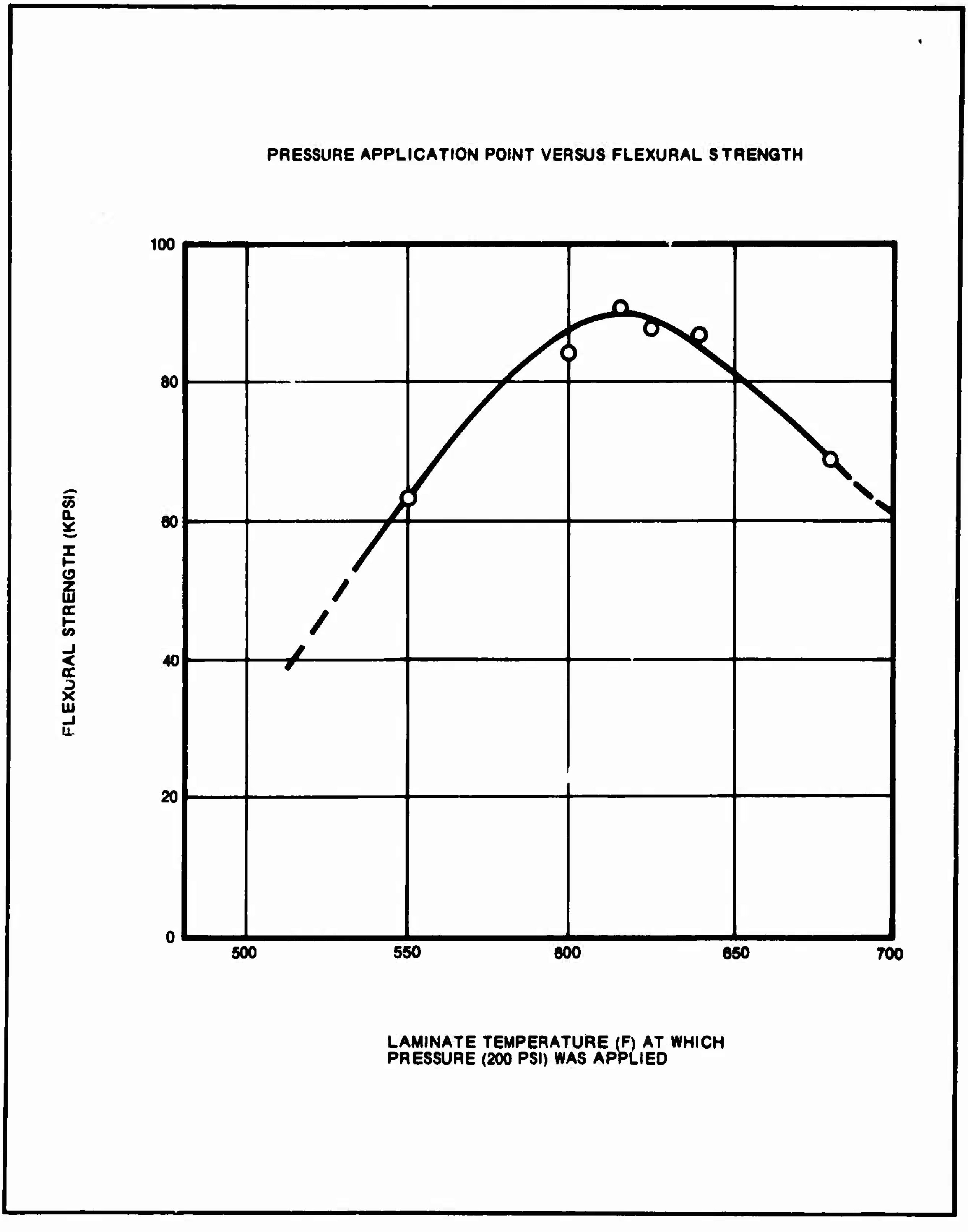



Figure 13 - Press Fabrication of 4-ply Laminate, PBI 2801 (AFR-100)

## TABLE XX

**AUTOCLAVE FABRICATION OF AFR-100 (2801 or Imitite 1850) PBI LAMINATES**

| SAMPLE | VAC. | PRESS PSI | THICKNESS INCHES | R.C. % | FLEXURAL STRENGTH PSI |
|---|---|---|---|---|---|
| 56* | Yes | No | .073 | 38.3 | 13,872 |
| 59 | Yes | 110 | .039 | 27.6 | 108,262 |
| 63 | Yes | No | .043 | 27.0 | 86,080 |
| 77 | Yes | No | .045 | 29.4 | 61,280 |
| 78 | Yes | 100 | .038 | 29.4 | 132,180 |
| 80 | Yes | 50 | .040 | 27.8 | 94,420 |

**NOTES:**

* 1. Sample #56 developed a leak in the bag during cure. The laminate was badly oxidized.

2. All laminates were 4-ply construction.

The following technique for bagging PBI resin systems for autoclave curing was developed and proved quite successful.

1. Five mil soft aluminum foil was used for the bag.

2. The lay-up was completely enveloped - top and bottom - in the foil bag.

3. A 1-1/2 to 2-inch wide strip of partially cured 12 mil silicone rubber tape was placed around the edge between the two foil layers.

4. The foil layers were crimped together by folding several times with the silicone tape sandwiched between.

5. The vacuum outlets were then sealed by means of solid silicone rubber gaskets and a threaded washer.

Figure 14 shows a sketch of the bagging method described above.

## C. PRESS FABRICATION OF PBI (AFR-151) FACE SHEETS

The information derived during the press processing of PBI 2801 was applied successfully to the processing of the PBI 2803 (AFR-151) prepreg. Table XXI presents the results of the process study on PBI 2803 (AFR-151) using the press fabrication method.

Figure 15, based on Table XXI, shows that the pressure-point technique applied. It should be noted that the optimum temperature range for applying pressure was nearly 50F higher for the AFR-151 resin than for the AFR-100 resin. (Compare Figures 15 and 13). The effect of pressure on the properties of press-cured PBI (2803 AFR-151) face sheet laminates is also shown in the data of Table XXI. It was apparent that a pressure below 200 psi was detrimental to the physical properties. However, there appeared to be little difference in the effect of pressure between 200 psi and 1000 psi as long as the pressure point cure technique was effectively applied. This corresponded to the information determined on the press curing of polyimide laminates as reported in Section III-B-2 of this report.

Based on the press fabrication study it was determined that the recommended processing for four-ply PBI (AFR-151) face sheets by the press method was:

1. Use 4-plies of 181 glass fabric reinforced PBI prepreg.

2. Use one-ply of 181 glass fabric release cloth on the surface eventually to be bonded to the honeycomb core.

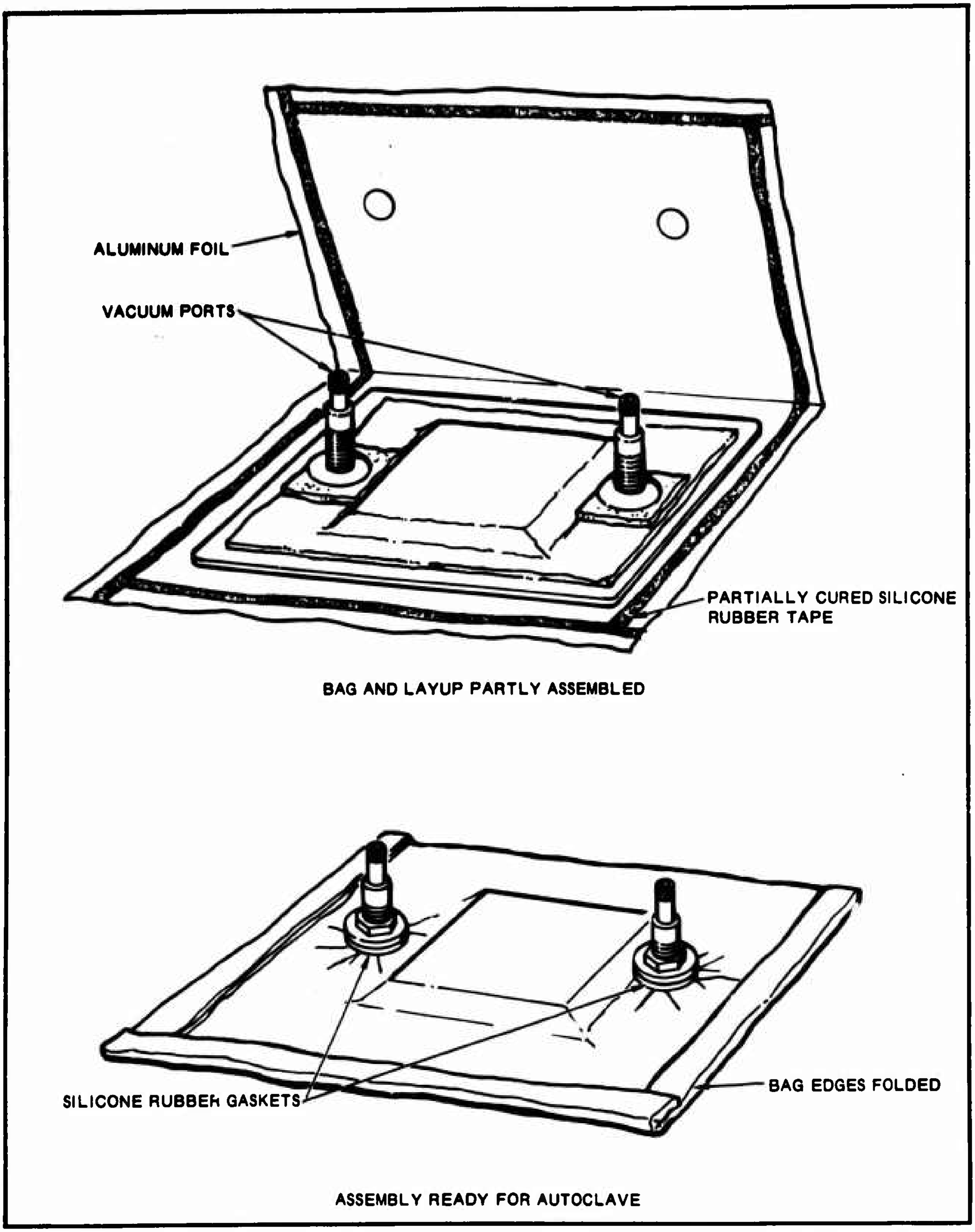



**Figure 14 - Bagging Technique for High-temperature Curing Autoclave**

## TABLE XXI

### PRESS FABRICATION OF AFR-151(2803) PBI LAMINATES

| Sample | Lam. Temp When Press. Was Applied Degrees F | Pressure PSI | Thickness Inches | R.C % | Flexural Strength PSI |
|---|---|---|---|---|---|
| 87 | 675 | 200 | .036 | 29.8 | 89,190 |
| 88 | 665 | 200 | .033 | 25.7 | 92,880 |
| 89* | 670 | 200 | .039 | 30.2 | 73,760 |
| 90 | 670 | 200 | .036 | 29.2 | 88,940 |
| 91 | 640 | 200 | .039 | 30.4 | 78,800 |
| 93 | 543 | 200 | .038 | 33.6 | 62,400 |
| 106 | 670 | 100 | .044 | 30.9 | 68,160 |
| 107 | 670 | 1000 | .030 | 30.7 | 87,740 |
| 111 | 680 | 200 | .040 | 35.8 | 82,260 |
| 112 | 680 | 200 | .041 | 27.1 | 73,900 |
| 113 | 675 | 200 | .034 | 22.0 | 82,100 |
| 114 | 675 | 200 | .040 | 23.3 | 77,400 |
| 115 | 675 | 200 | .044 | 32.9 | 72,770 |

NOTES:

* 1. Photo cell on press instrument panel failed. Temperature readings were questionable. Sample 90 was a repeat of Sample 89.

2. All laminates were 4-ply construction.

3. Pressure was 200 psi.

4. The initial press temperature was 350 F.

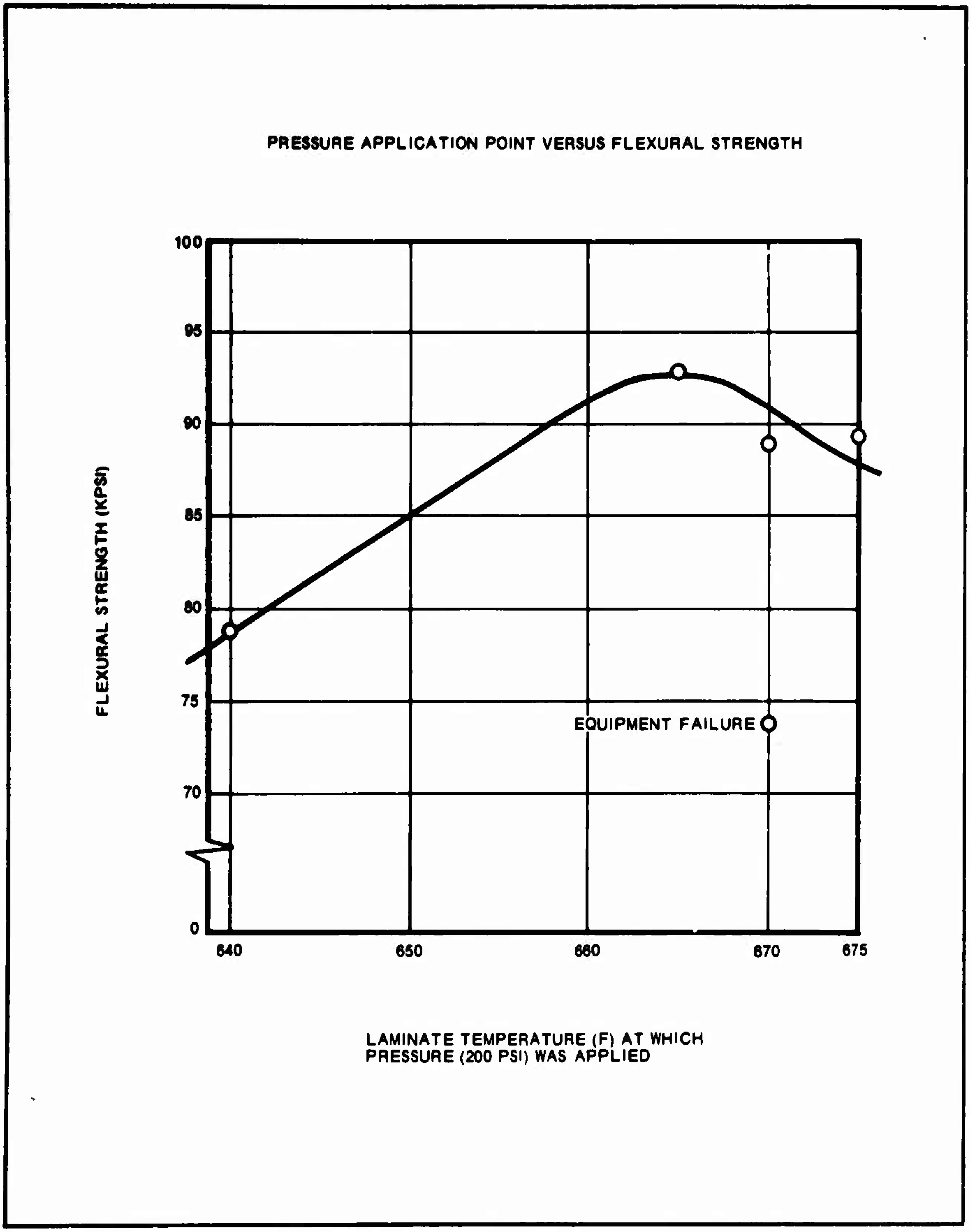



Figure 15 - Press Fabrication of 4-ply Laminate, PBI 2803 (AFR-151)

3. Place the lay-up between aluminum caul plates treated with a Ram GS-3 release agent.

4. Initial press temperature should be 350F.

5. Use shims that will allow the press platens to close within approximately .030 of the lay-up without exerting pressure on the part.

6. Set the press temperature to 700F after the lay-up has been inserted.

7. Remove shims and pressure the lay-up to 200 PSI at a predetermined part temperature. The temperature is read from a thermocouple in the part. The pressure-point may vary depending on press heat-up rate, prepreg characteristics, and resin formulation. This pressure-point must be determined for each specific set of conditions, but once determined will remain the same as long as the conditions remain the same.

8. Cure the laminate for 90 minutes from the time of pressure application.

9. Cool under pressure to 350F.

10. Do not remove the release cloth peel ply until just prior to bonding into a sandwich composite.

11. Post-cure in an inert atmosphere as follows: 8 hours at 400F, 8 hours at 450F, 8 hours at 500F, 3 hours at 600F, 5 hours at 700F, 1 hour at 800F, and 1 hour at 850F.

## D. AUTOCLAVE FABRICATION OF PBI (AFR-151) FACE SHEETS

The information developed during the autoclave processing of PBI 2801 (AFR-100) was not applied as successfully to PBI 2803 (AFR-151) as had been the case in the development of press fabricating techniques. The PBI 2803 resin required higher curing temperatures and exhibited greater fluidity during conversion. These two factors of resin fluidity and high cure temperature made autoclave processing of thin face sheet laminates extremely difficult.

Table XXII presents data on the laminates prepared during the autoclave processing study on PBI 2803. The intent of this processing study was not only to obtain sandwich face sheet laminates of peak quality but to do so, if at all possible, by fabrication techniques that were compatible with standard production facilities and methods. For this reason an intense effort was made to determine a satisfactory autoclave procedure that did not require excessively high pressures and temperatures or an

## TABLE XXII

**AUTOCLAVE FABRICATION OF AFR-151(2803) PBI LAMINATES**

| Sample | Vacuum | Pressure PSI | Type of Cure | Lam Temp when Press was Applied °F. | Elapsed Time Before Pressuring Mins. | Rate of Heating °F/Min | Thickness Inches | Before Post-Cure | | After Post-Cure | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | Resin Content | Flexural Strength | Resin Content | Flexural Strength |
| 94(1) | Yes | 100 | Press. Pt. | 425 | 135 | - | | | N.C. | | |
| 95 | Yes | 100 | Step-cure | 425 | 120 | 2.9 | .030 | 17.6 | 26,000 | 17.5 | 87,000 |
| 96 | Yes | 100 | Step | 450 | 113 | 3.3 | .030 | 17.6 | 38,467 | 18.6 | 76,900 |
| 97 | Yes | 100 | Step | 475 | 117 | 3.4 | .030 | 18.1 | 55,800 | | |
| 98 | Yes | 100 | Step | 490 | 170 | 2.4 | .033 | 23.4 | 28,333 | | |
| 108(2) | Yes | None | Press. | - | - | - | | | N.G. | | |
| 109(3) | Vent(4) | 100 | Press. | 498 | 165 | 2.5 | | | N.G. | | |
| 110(5) | Vented | 100 | Step | 475 | 208 | 1.9 | .035 | 24.3 | 31,580 | 24.4 | 86,840 |
| 119 | Vented | 100 | Press. | 640 | 65 | 9.1 | .030 | 14.3 | 59,220 | 16.4 | 88,600 |
| 120 | Vented | 100 | Press. | 570 | 85 | 5.7 | .032 | 22.1 | 72,660 | 21.1 | 82,820 |
| 121 | Vented | 100 | Press. | 575 | 100 | 4.95 | .033 | 23.6 | 66,220 | 23.3 | 81,380 |
| 122 | Vented | 100 | Step | 460 | 188 | 2.0 | .032 | | | 21.1 | 78,433 |

Notes:

1. Leak in vacuum bag. Part oxidized.
2. Vacuum only during processing. No interlaminar bond.
3. Gellation had occurred before pressure was applied.
4. Samples 109, 110, 119, 120, 121, and 122 were vented to the atmosphere and held under 5 psi pressure during the initial heat-up period.
5. A portion of sample 110 was post-cured in a press at 425 PSI and 700F for 3 hours. Flexural strength after post-cure was 91,520 PSI.

extremely long cure cycle. As the temperature, pressure, and cycle time requirements for autoclaves increase, the cost of production facilities and the cost of operations rise sharply.

An analysis of the data collected during the autoclave fabrication study (Table XXII) indicated the following:

1. The fluid nature of the resin during the conversion stage made it impractical to maintain a vacuum on the part during the entire cure cycle. The vacuum would cause excessive resin flow into the bleeder cloth resulting in a resin-starved laminate. Venting the laminate to the atmosphere and maintaining a slight autoclave pressure (5 psi) during the initial portion of the cure cycle proved satisfactory.

2. The higher cure temperature coupled with the restricted pressures and temperatures imposed by the autoclave made a post cure treatment mandatory.

3. Pressure was necessary during the latter portion of the autoclave cure in order to produce acceptable PBI 2803 laminates. An autoclave pressure of 100 psi was satisfactory.

4. The autoclave cure temperature could be kept below 550F provided an extended step-cure cycle was employed. In this technique the laminate was vented to the atmosphere and the autoclave maintained at 5 psi during the initial portion of the cure. The cure then proceeded as follows:

    90 Minutes at 350F (autoclave temp)
    30 Minutes at 400F
    20 Minutes at 450F
    (Pressure autoclave to 100 psi when temperature reached 475).
    180 Minutes at 500F

    The laminate was cooled to below 200F under pressure. It was discovered that the laminate possessed less than 50% of its ultimate strength at this stage and had to be handled carefully to prevent delamination.

    Ultimate strength was attained by post-curing the laminate in an inert atmosphere according to the following schedule:

    8 hours at 400F
    8 hours at 450F
    8 hours at 500F
    3 hours at 600F

5 hours at 700F
1 hour at 800F
1 hour at 850F

5. The autoclave cure cycle could be shortened by using the pressure-point technique rather than the step-cure method. An autoclave temperature in excess of 600F was required for the pressure-point system, however. It became apparent that the conversion characteristics of PBI (2803) were such that restrictive compromises were necessary for autoclave fabrication of thin laminates. If autoclave temperatures were to be restricted to the low 500F temperature range then an extended step-wise cure method had to be employed. If it was desirable to employ the more efficient pressure-point technique, then it was necessary to attain a laminate temperature in excess of 600F at a heating rate of approximately 5F/minute.

Based on the autoclave fabrication study, the recommended processing for four-ply PBI 2803 (AFR-151) face sheets by the autoclave method was determined to be:

1. Position 4-plies of 181 glass cloth reinforced PBI prepreg and one ply 181 release cloth on an aluminum base plate.

2. Cover the prepreg with a perforated teflon barrier film.

3. Cover the barrier film with several plies of bleeder cloth.

4. Use an edge bleeder around the periphery of the lay-up.

5. Position several vacuum outlets (never less than 2) on the edge bleeder.

6. Use a 5 mil soft aluminum foil envelope.

7. Seal the foil with uncured silicone tape.

8. Pull a vacuum on part and check for leaks.

9. Insert part in autoclave. Pressurizing to 5 PSI, then shut-off vacuum and vent to atmosphere. Either of the following cure cycles could then be used.

   a. Pressure-point method of cure:

      1) Raise autoclave temperature from R. T. to 620F at a rate of approximately 5F/minute.

2) When part temperature (thermocouple) reached a predetermined value, increase pressure to 100 PSI.

b. Step method of cure.

1) Raise autoclave temperature from R. T. to 350F at a rate of approximately 8F/minute. Hold at 350F for 90 minutes.

2) Raise autoclave temperature to 400F at a rate of 5F/min. Hold at 400F for 30 minutes.

3) Raise autoclave temperature to 450F and hold for a total time of 20 minutes.

4) Raise autoclave temperature to 500F. When part reached 475F (thermocouple) increase pressure to 100 PSI.

10. Cure for an additional 2-3 hours after pressurizing.

11. Cool to below 300F under pressure before removing from autoclave.

12. Post-cure in inert atmosphere as follows: 8 hours at 400F, 8 hours at 450F, 8 hours at 500F, 3 hours at 600F, 5 hours at 700F, 1 hour at 800F, and 1 hour at 850F.

13. Remove release cloth peel ply just prior to bonding into a sandwich composite.

## SECTION VI

## PROCESS STUDY - POLYBENZIMIDAZOLE SANDWICH COMPOSITES

### A. GENERAL

The supplier of PBI preimpregnants found that the AFR-151 (2803) resin produced an adhesive film which was too brittle and difficult to handle to be practical for use in sandwich composites. It was recommended that AFR-100 (2801) resin be used for the adhesive film. Compatibility of the two resins was considered excellent and no difficulty was anticipated in using 2801 resin adhesive to bond together skins and core containing 2803 resin.

The change to the AFR-100(2801) resin for the adhesive film was approved by the Applications Division, Air Force Materials Laboratory.

The materials used in the fabrication study on PBI 2803(AFR-151) sandwich composites were:

1. Face Sheets - As mentioned in Section V, the prepreg stock consisting of AFR-151 (2803) resin on 181 E-glass reinforcement was obtained from the Narmco Materials Division, Whittaker Corp., under the designation: "PBI 2803-1581 E-glass".

2. Honeycomb Core - The core stock was procured from Hexcel Products, Inc. The core resin was AFR-151 (PBI) and the reinforcement was E-glass fabric. Core densities were 4 pounds and 8 pounds per cubic foot. Core thickness was one-half inch and the cell size was three-sixteenth-inch.

   The core materials were procured under the following designations: "HRH-325(AFR-151 PBI Resin), 3/16 cell, 4.0 lb. density, .500" thick" and "HRH-325 (AFR-151 PBI Resin) 3/16 cell, 8.0 lb. density, .500" thick".

3. Adhesive - The adhesive was composed of AFR-100 (PBI) resin on HG32 style glass reinforced carrier. The adhesive was procured from the Narmco Materials Div., Whittaker., under the designation: "Imidite 850, HG32 Adhesive (PBI-2801)".

   The fabricating techniques evaluated in this study were:

   a. Secondary Bond

   b. Single Stage Operation

   c. Multiple Stage Operation

## B. SECONDARY BOND TECHNIQUE

Face sheet laminates evaluated in this secondary bond technique study were prepared by both the press method and the autoclave method.

Table XXIII presents the data on the PBI (2803) laminates used for the sandwich face sheets. It should be noted that the laminates prepared by the press method were not post-cured while the laminates prepared by the autoclave technique were all post-cured. PBI (2803) resin required a temperature in excess of 625F to effect complete conversion. This could be achieved in the press, but was somewhat impractical in the autoclave due to equipment restrictions and bagging limitations. The laminates cured in the autoclave were weak and subject to damage until after the postcure operation.

Both press method and the autoclave method were also employed to effect the face sheet to core bond.

1. Face sheets prepared by the press technique; secondary bonding of face sheets to core performed in the press. (P/P)

2. Face sheets prepared by the press technique; secondary bonding of face sheets to core performed in the autoclave. (P/AC)

3. Face sheets prepared by the autoclave technique; secondary bonding performed in the autoclave. (AC/AC)

4. Face sheets prepared by the autoclave technique; secondary bonding performed in the press. (AC/P)

In the press method for making secondary bonds, machined stops were used to prevent crushing the core. See Figure 16. The cure cycle for preparing secondary bonds by the press method was as follows.

1. Preheat press to 430F.

2. One hour at 430F.

3. One hour at 600F.

4. Cool to 350F before release.

In the autoclave method (Figure 17) for making secondary bonds the special sandwich panel fixture (Figure 7) was used. Early in the processing study it was found that the autoclave pressure had to be limited to 30 psi. Pressures in excess of 30 psi crushed the PBI honeycomb core during the cure cycle. This is illustrated by Figure 18. The top panel shows a sandwich composite subjected to 50 psi during secondary

## TABLE XXIII

**PBI(2803) Laminates Prepared for Use as Face Sheets on Secondary Bonded Sandwich Composites**

| Sample | Fabrication Method | Thickness | R. C. % | Flexural Strength PSI |
|---|---|---|---|---|
| 111 | Press | .040 | 35.8 | 82,260 |
| 112 | Press | .041 | 27.1 | 73,900 |
| 113 | Press | .034 | 22.0 | 82,100 |
| 114 | Press | .040 | 23.3 | 77,400 |
| 115 | Press | .044 | 32.9 | 72,770 |
| 121 | Autoclave | .033 | 23.3 | 81,380 (after post cure) |
| 122 | Autoclave | .032 | 21.1 | 78,433 (after postcure) |
| 123 | Autoclave | .031 | 19.5 | 98,440 (after postcure) |

Notes:

1. A ply of release cloth was laminated to one surface of each panel to serve as a peel ply.

2. Samples 122 and 123 were postcured in an inert atmosphere according to the following schedule:

   8 hours at 400 F
   8 hours at 450 F
   8 hours at 500 F
   3 hours at 600 F
   5 hours at 700 F
   1 hour at 800 F
   1 hour at 850 F

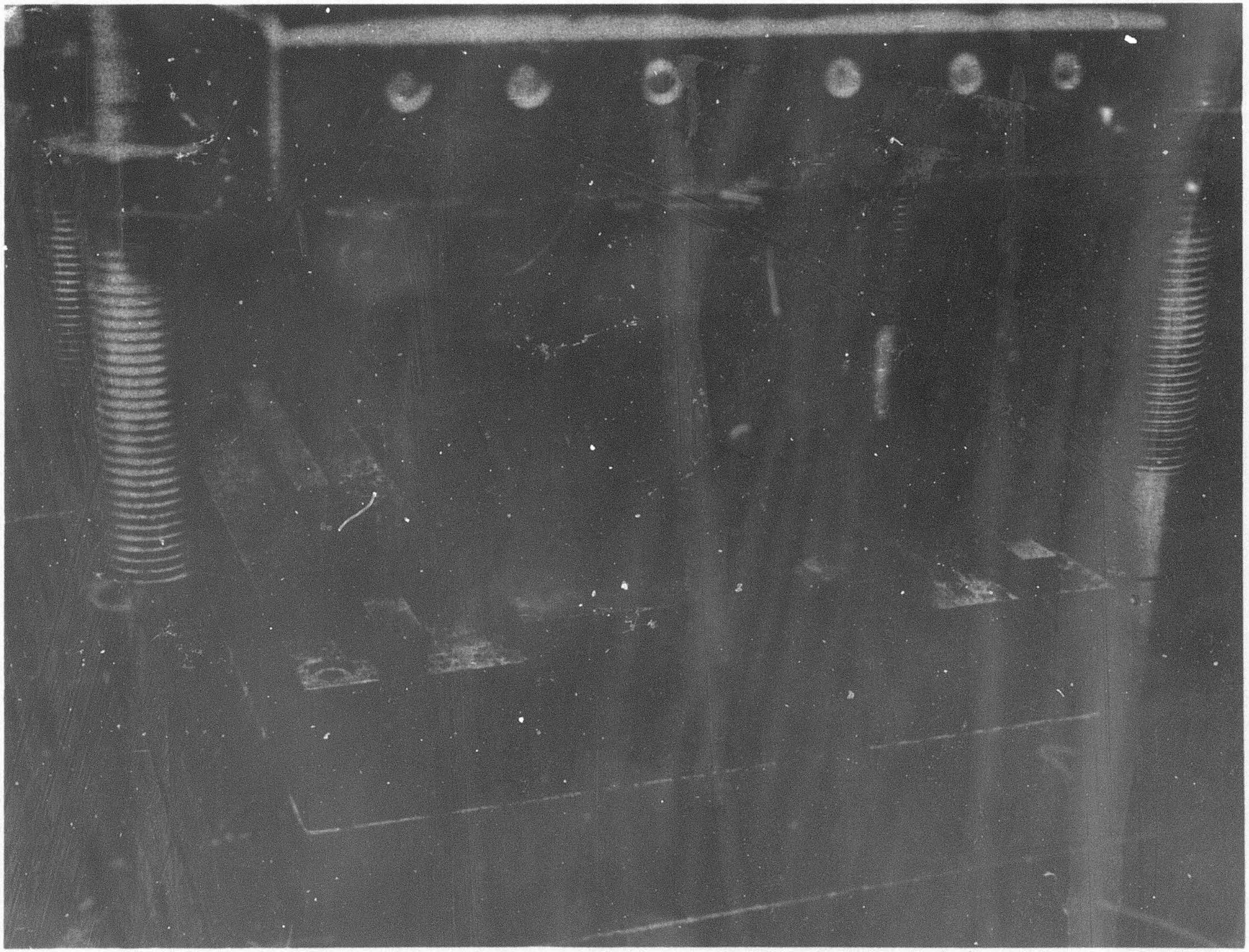


Figure 16 - Secondary Bonding by the Press Technique

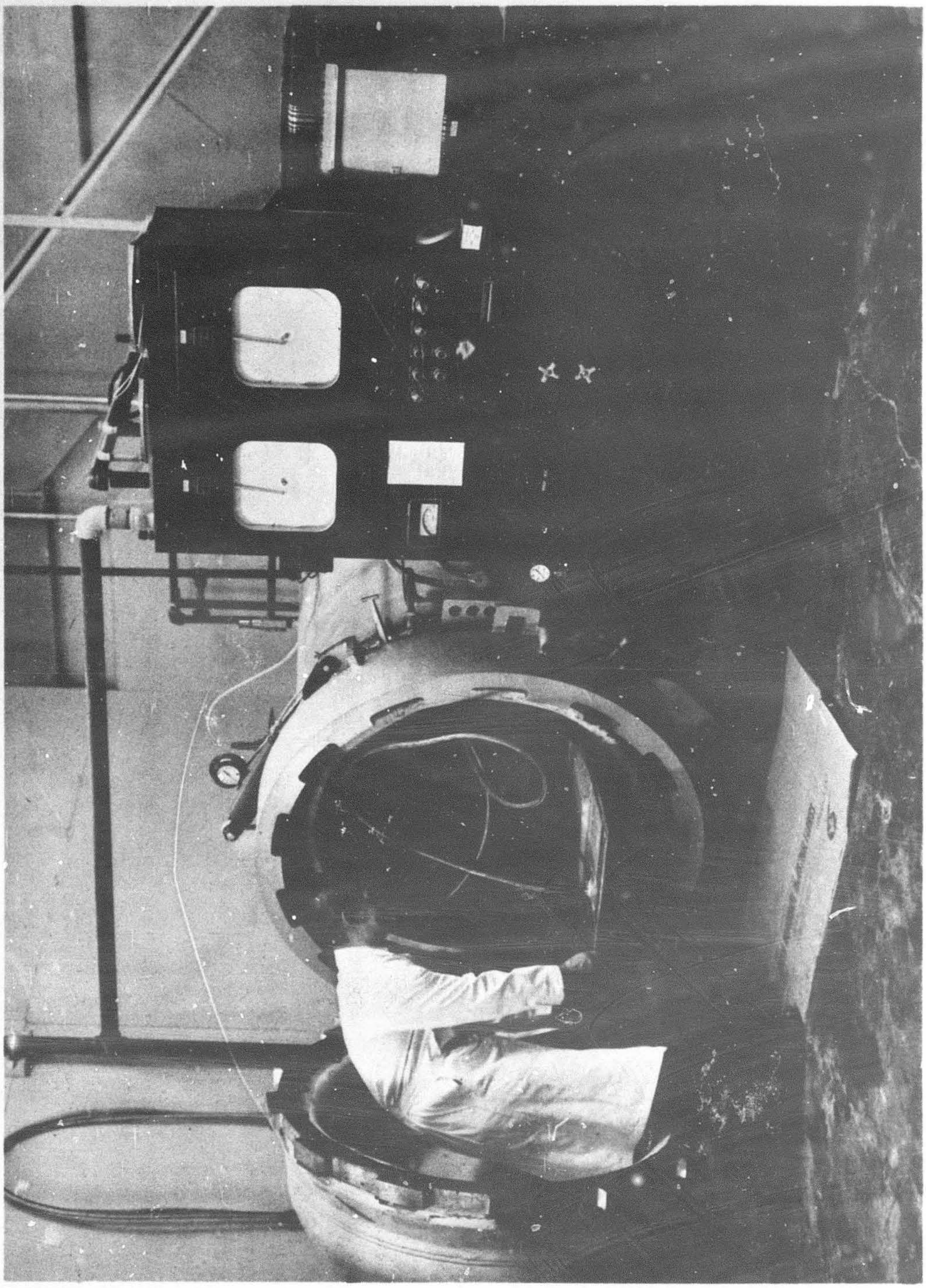
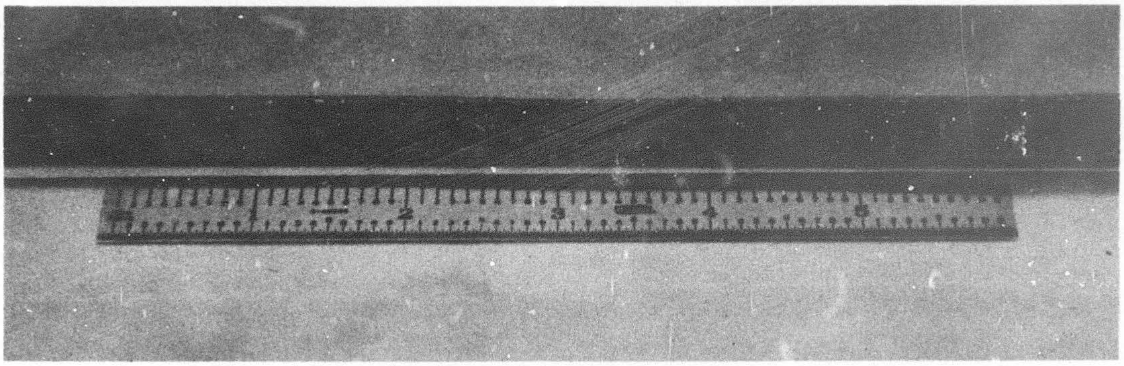
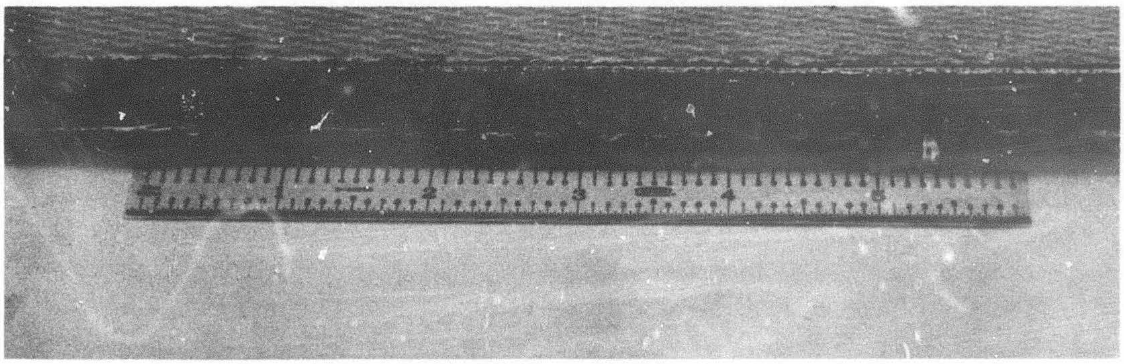


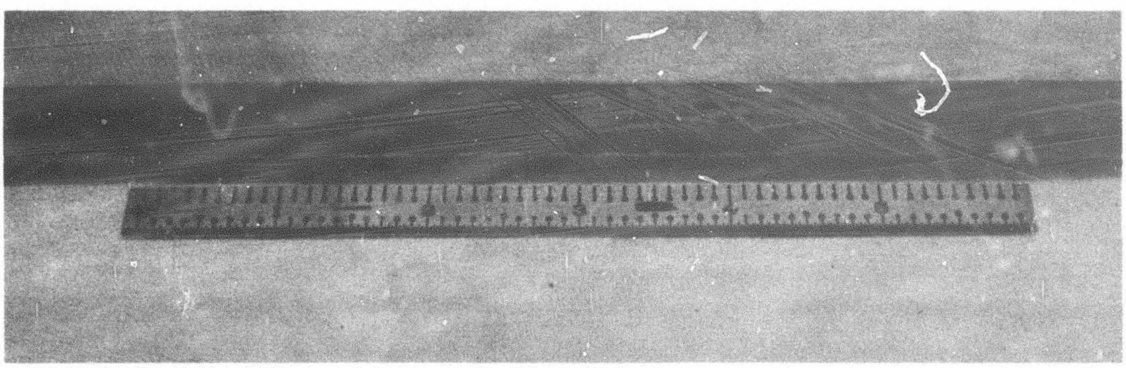
Figure 17 - High-Temperature, High-Pressure Autoclave Facility



CRUSHED PBI CORE - AUTOCLAVE PRESSURE - 50 PSI



ACCEPTABLE SANDWICH COMPOSITE - AUTOCLAVE PRESSURE - 30 PSI



PRESS CURED SECONDARY BOND. PRESS CLOSED TO STOPS.

Figure 18 - Example of Core Crushing

bonding. The middle panel shows a composite prepared at 30 psi autoclave pressure. The lower panel shows a composite where the secondary bond was prepared in a press using stops.

As the processing study continued, the critical aspect of the pressure became more and more apparent. Pressures were lowered to 25 psi and finally to 20 psi to prevent any tendency toward core collapse during the cure cycle.

The autoclave cure cycle for preparing secondary bonds was:

1. Bagged assembly was placed in room temperature autoclave and pressured to 30 psi. (Part was vented to atmosphere).

2. Autoclave heated to 430F.

3. One hour at 430F.

4. One hour at 600F.

5. Cool to 350F.

Table XXIV presents the data from the secondary bond study on PBI (2803) sandwich composites.

## C. SINGLE STAGE OPERATION

Three sets of conditions were evaluated in the single-stage technique.

1. Curing by means of the pressure-point technique.

2. Curing by means of the step method.

3. Omitting the adhesive films and using the resin in the face sheet prepreg plies to bond the face sheets to the core.

The PBI sandwich composites were assembled as shown in Figure 19. The four-plies of prepreg for the face sheets, the adhesive films, and the core were stacked, with the adhesive films between the face sheets and the core. This assembly was placed in the special sandwich panel fixture.

The procedure used in preparing the single-stage PBI sandwich composites was as follows. See Figure 20.

1. A 5 mil soft aluminum foil was used as the bagging envelope. Strips of uncured silicone tape were laid along the edge of the foil to serve as a sealant. The foil in this area was coated with a silicone primer to improve the bond of the tape to the foil.

## TABLE XXIV

## SECONDARY BOND

### PBI(2803) Honeycomb Core Sandwich Composites

| Composite No. | Face Sheet Process | Ahesive Cure Process | Flexural Strength lbs/in Width | Core Shear | Flatwise Tensile |
|---|---|---|---|---|---|
| 29 | Press | Press | 241 | 224 | 329 |
| 48 | Autoclave | Autoclave | 170.5 | 159.5 | 424 |
| 49 | Autoclave | Press | 158.3 | 148 | 414 |
| 53 | Press | Autoclave | 168.7 | 158 | 418 |

Notes:

1. Machined stops were used when secondary bonds were made in press.
2. A pressure of 30 psi was used when secondary bonds were made in the autoclave.
3. Face sheet laminates were not post cured prior to secondary bonding.
4. Sandwich composites were postcured before testing.

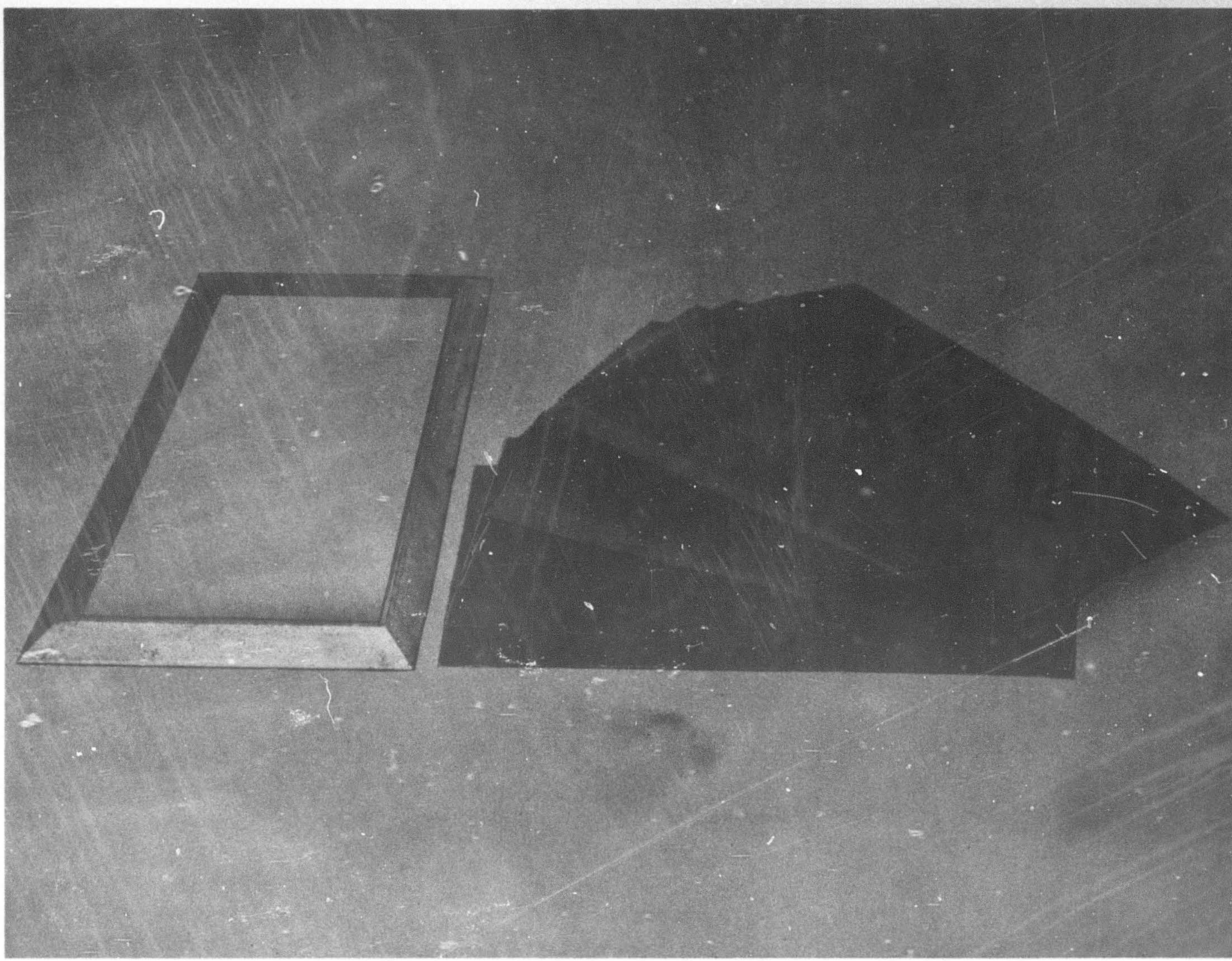


Figure 19 - Sandwich Layups for Single Stage Cure

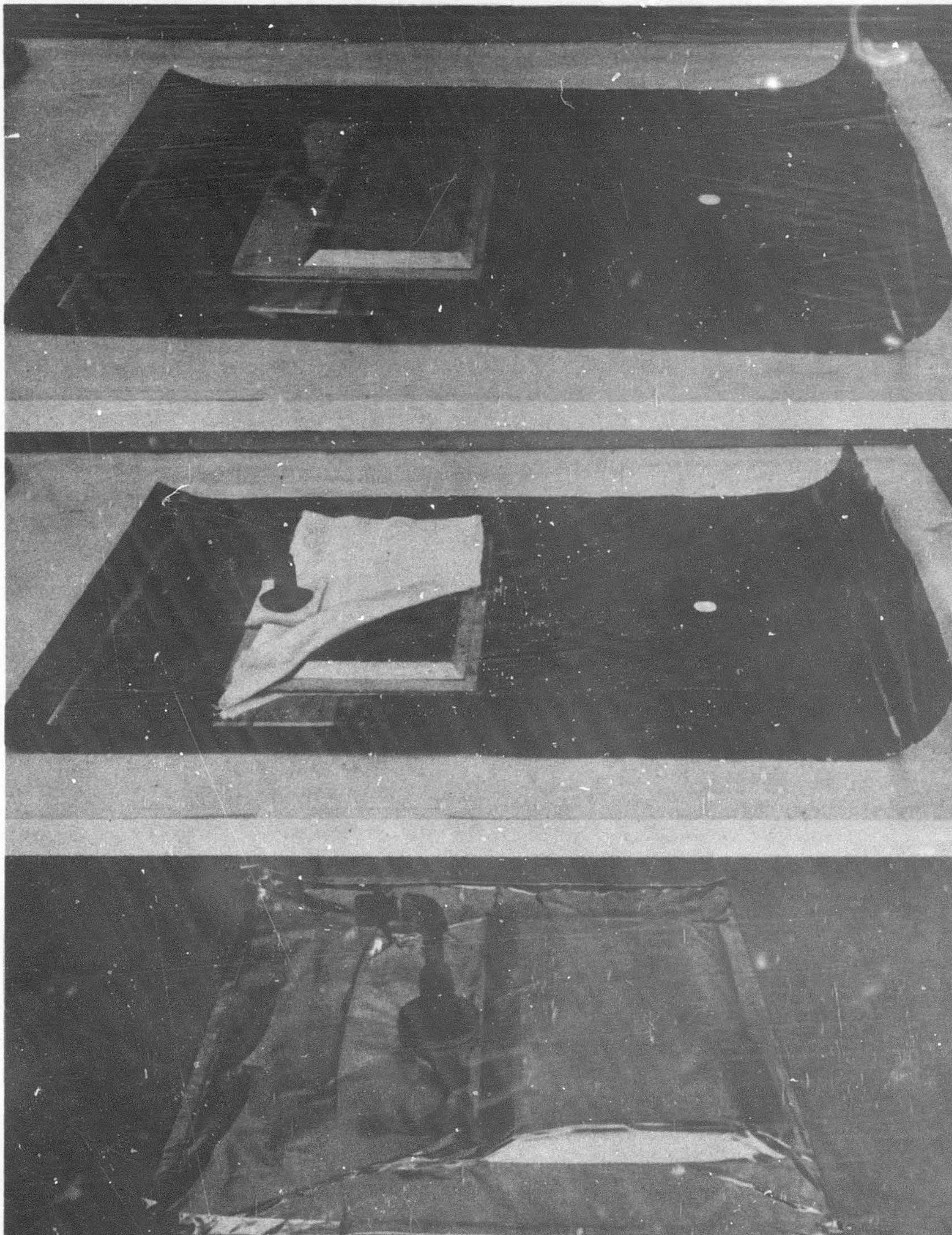


Figure 20 - Layup Technique for PBI Sandwich Composite

2. An aluminum base plate was placed on the foil and the special frame then positioned on the base plate.

3. Four plies of PBI (2803) prepreg (lower face sheet lay-up) were placed in the special frame.

4. One ply of PBI (2801) adhesive - Imidite 850 - was placed on the prepreg:

   NOTE: This step was omitted, of course, in the composite made without using an adhesive film.

5. The PBI honeycomb core (HRH-325) was positioned on the adhesive film.

6. A ply of PBI adhesive was placed on the core.

   NOTE: This step was omitted, of course, in the composite made without using an adhesive film.

7. Four plies of PBI prepreg (upper face sheet lay-up) were positioned on the adhesive film.

8. A thermocouple junction was placed between the third and fourth plies at the edge of the top face sheet lay-up.

9. The entire composite was covered with a perforated teflon barrier film.

10. The barrier film was covered with several plies of bleeder cloth. The cloth was allowed to overlap the frame an amount sufficient to accommodate a vacuum port.

11. A vacuum port was positioned on the bleeder cloth at the edge of the frame holding the composite lay-up.

12. The aluminum foil was folded over the composite lay-up. A hole was cut in the foil bag to accommodate the vacuum port. Silicone rubber gaskets were used to seal around the port.

13. The aluminum foil envelope was sealed together with the uncured silicone tape. The use of the silicone primer assured the bond of the tape to the foil.

14. The edges of the foil envelope were crimped together to assure a tight seal.

15. A vacuum was pulled on the part and a check made for leaks - especially at the point where the thermocouple lead wires emerge.

16. The assembly was inserted in the autoclave and pressurized to 5 psi. The vacuum was then shut off and the part vented to the atmosphere.

17. The next portion of the cycle depended on whether the pressure point cure technique or the step cure technique was employed.

    a. Pressure point technique

        1) The autoclave temperature was raised from R. T. to 620F at a rate of approximately 5F/minute.

        2) When part temperature (thermocouple) reached a predetermined value, the pressure was increased to 30 psi.

    b. Step method of cure

        1) The autoclave temperature was raised from R. T. to 350F at a rate of approximately 8F/minute and then held at 350F for 90 minutes.

        2) The autoclave temperature was raised to 400F at a rate of 5F/minute and held at 400F for 30 minutes.

        3) The autoclave temperature was raised to 450F and held for a total time of 20 minutes.

        4) The autoclave temperature was raised to 500F. When the part reached 475F (thermocouple), the pressure was increased to 30 psi.

18. The composite was cured for an additional 2-3 hours after pressurizing.

19. The composite was cooled to below 300F under pressure before removing from autoclave.

20. Post curing was performed in an inert atmosphere as follows: 8 hours at 400F, 8 hours at 450F, 8 hours at 500F, 3 hours at 600F, 5 hours at 700F, 1 hour at 800F, and 1 hour at 850F.

Table XXV presents data from the single-stage study on PBI (2803) sandwich composites.

## TABLE XXV

## SINGLE STAGE

### PBI(2803) Honeycomb Core Sandwich Composites

| Composite No. | Process | Flexural Strength lbs/in Width | Core Shear | Flatwise Tensile |
|---|---|---|---|---|
| 35 | Step Cure | 251 | 235 | 350 |
| 39 | Pressure Point | 287 | 268 | 337 |
| 40 | No Adhesive (Step Cure) | 146.2 | 136.8 | 98 |
| 43 | Step Cure | 192.2 | 187.4 | 493 |
| 46 | Pressure Point | 331 | 309 | 359 |
| 60 | Pressure Point | 336 | 313 | 190 |
| 61 | Step Cure | 223 | 207 | 211 |
| 62 | Step Cure | 228 | 213 | 202 |

Notes:

1. Autoclave pressure during cure was 30 psi.
2. Sandwich composites were postcured before testing.

## D. MULTIPLE STAGE OPERATION

The multiple-stage technique employed in the processing study on PBI (2803) sandwich composites was the "Three-Stage Method". In the initial stage of this technique the outer skin (with release cloth peel ply) was laid-up and cured in the autoclave in the normal manner for skin laminates. In the second stage the peel ply was removed from the cured skin laminate and the core was bonded to the exposed surface by means of a film adhesive. In the third stage, a layer of adhesive was laid over the bonded core, skin prepreg plies were added, the assembly was covered with barrier film, bleeder cloth and sealed bag, and cure performed in an autoclave using the pressure-point technique.

As discussed in Section IV-E, the multiple stage technique can have two advantages in some Production situations.

1. The outer surface skin can be laminated at pressure well above the crushing point of the honeycomb core. This enhances the possibilities of a dense, void-free surface skin.

2. In many cases the shape of an aerospace vehicle component is such that assembly of the complete sandwich composite for a single-stage cure is difficult or impractical.

The multiple stage technique was evaluated in this program on PBI (2803) sandwich composites as a feasibility study only - to determine the feasibility of the process and the effect on physical properties of the composite.

Table XXVI presents some typical physical properties data on PBI (2803) sandwich composites fabricated by several processes. The data indicated that the multiple stage technique was feasible and that sandwich composites possessing acceptable properties could be fabricated by that method.

## E. DISCUSSION ON THE PROCESSING OF PBI(2803) SANDWICH COMPOSITES.

The fabrication of PBI (2803) honeycomb core sandwich composites presented difficulties. An analysis of the data in Tables XXIV and XXV showed that, while the strength levels were generally quite good, there was considerable variation. The variation did not seem to form a predictable pattern.

The cause of the variations could be ascribed to one or more of the following factors.

TABLE XXVI

Comparison of Process Methods for

PBI(2803) Sandwich Composites

| Composite No. | PROCESS | Flexural Strength Lbs/In Width | Flexural Strength Core Shear | Flatwise Tensile |
|---|---|---|---|---|
| 29 | Secondary Bond - Press | 241 | 224 | 329 |
| 48 | Secondary Bond - Autoclave | 171 | 160 | 424 |
| 46 | Single Stage - Pressure Point | 331 | 309 | 359 |
| 61 | Single Stage - Step Cure | 223 | 207 | 211 |
| 79 | Multiple Stage - Three Step Cure | 253 | 237 | 250 |

1. Variations in the prepreg. - There were some obvious variations in the PBI prepreg used in this contract. The prepreg was prepared by a hot melt technique which is inherently subject to variations in coating weight, thickness, volatile content, reinforcement penetration, and balance. An attempt was made during preparation of test panels to discard all prepreg areas with heavy or light resin content.

   The prepreg was also quite dry and brittle. Even with careful handling some loss of resin occurred by crumbling and flaking during the cutting and lay-up operations.

2. Variations in adhesive film. - The same problems existed with the adhesive film as were discussed in connection with the prepreg. An additional variation lay in the fact that the adhesive resin was AFR-100 (2801) resin while the prepreg resin was AFR-151 (2803) resin. These two resins were compatible but the flow properties, outgassing, and gelation temperature were different.

3. Variations in the honeycomb core. The PBI honeycomb core was hand made by a laboratory process. This was subject to the normal human variances.

4. Process conditions. - The high-temperature and long curing cycles required for PBI (2803) put an abnormal strain on lay-up techniques, bagging film, sealants, thermocouples, autoclave connections, etc.

5. Oxidation. - The PBI resins were extremely sensitive to oxidation, especially at high-temperatures and before the resin was completely cured. If the encasing bag developed the slightest leak during an autoclave cure, the PBI part being cured was subject to degradation. The condition was even more extreme during postcure because temperatures were carried as high as 850F. (This is discussed in greater detail in the following section, VI-F.)

6. Bondable surface preparation for secondary bonding. - The use of a peel ply to provide a bondable surface presented problems. The fluidity of the resin, the high-temperature involved, and the unique adhesiveness of the resin caused the peel ply to adhere tenaciously to the laminate. The peel ply could be removed by sand blasting (Sample 29, Table XXIV) but this provided a less than optimum bonding surface.

   In Sample 53, Table XXIV, the press cure cycle was reduced. This permitted the peel ply to be removed in normal fashion. The face sheet laminate was then post cured to achieve

desired physical properties. Difficulty with the peel ply was not experienced in autoclave cured face sheet laminates because cure conditions were less extreme than with the press method.

7. **Pressure.** - It was found that the autoclave pressure had to be limited to 20 psi during the cure cycle on PBI honeycomb core sandwich panels. A pressure of 50 psi was preferred when curing PBI sandwich composites, but core crushing occurred at the higher pressure.

After analysing the results of the study on PBI (2803) honeycomb sandwich composites, it was decided to prepare the heat aging specimens for Phase II by means of the single-stage technique. This decision was based on observations, handling considerations, and production-oriented concerns, as well as on the physical properties data. The following factors influenced the decision.

1. The highest physical properties were obtained by the single-stage technique.

2. The single-stage technique required less time per panel than the secondary bond method. In fact, the secondary bonded panels required nearly twice as long to fabricate. The face sheets had to be cured and then bonded to the core. The face sheet laminating cycle and the bonding cycle were about equal in length. The single-stage method required only one cure cycle.

3. The two cure cycles in the secondary bond method increased the opportunities for processing difficulties to arise and for oxidation to occur. This could increase the chances for variations.

4. The problems involved in making an optimum bonding surface in the secondary bond method could create undesirable additional variations.

5. From a production processing point of view, the single-stage method was preferred.

## F. POST CURE OXIDATION STUDY

The post curing of PBI composites must always be performed under an inert atmosphere because the PBI resins are susceptible to oxidation. During the process development work on face sheet laminates, the postcure step was conducted in a stainless steel envelope under a blanket of nitrogen.

The schedule used for post curing PBI (2803) composites was:

8 hours at 400F
8 hours at 450F
8 hours at 500F
3 hours at 600F
5 hours at 700F
1 hour at 800F
1 hour at 850F

The appearance of the post cured laminates was excellent and physical properties high. (Table XXIII)

In order to accommodate the larger sandwich panels to be prepared for the heat aging study, a new heat aging chamber was designed and constructed (Figure 21). The chamber was made of welded stainless steel with a sealed door. The chamber was positioned in a large laboratory oven (Figure 22). Nitrogen was metered (see Figure 23) into the top of the chamber to create an inert atmosphere.

Several PBI (2803) laminates were post cured in the new chamber with disappointing results. (Table XXVII). The laminates were quite dark in color and the flexural strength values were lower than desired, indicating oxygen degradation.

An investigation was conducted to determine if oxygen degradation was occurring in the new post cure chamber. A laminate was prepared, cut into four sections, and conditioned as follows.

1. No post cure

2. Post cured in air

3. Post cured in our regular stainless steel envelope under nitrogen.

4. Post cured in the new chamber under nitrogen.

Table XXVIII presents the results of the post cure test. The detrimental effect of oxidation was readily apparent in the 75% loss of strength by the section post cured in air. The differences in strength between the sections post cured under nitrogen indicated that the envelope was considerably more efficient than the new chamber.

The new chamber was modified to prevent any air leakage around the door or at the seams and gas connections. A more extensive purging method was incorporated into the operating procedure. The use of Argon gas as the inert blanket was evaluated.

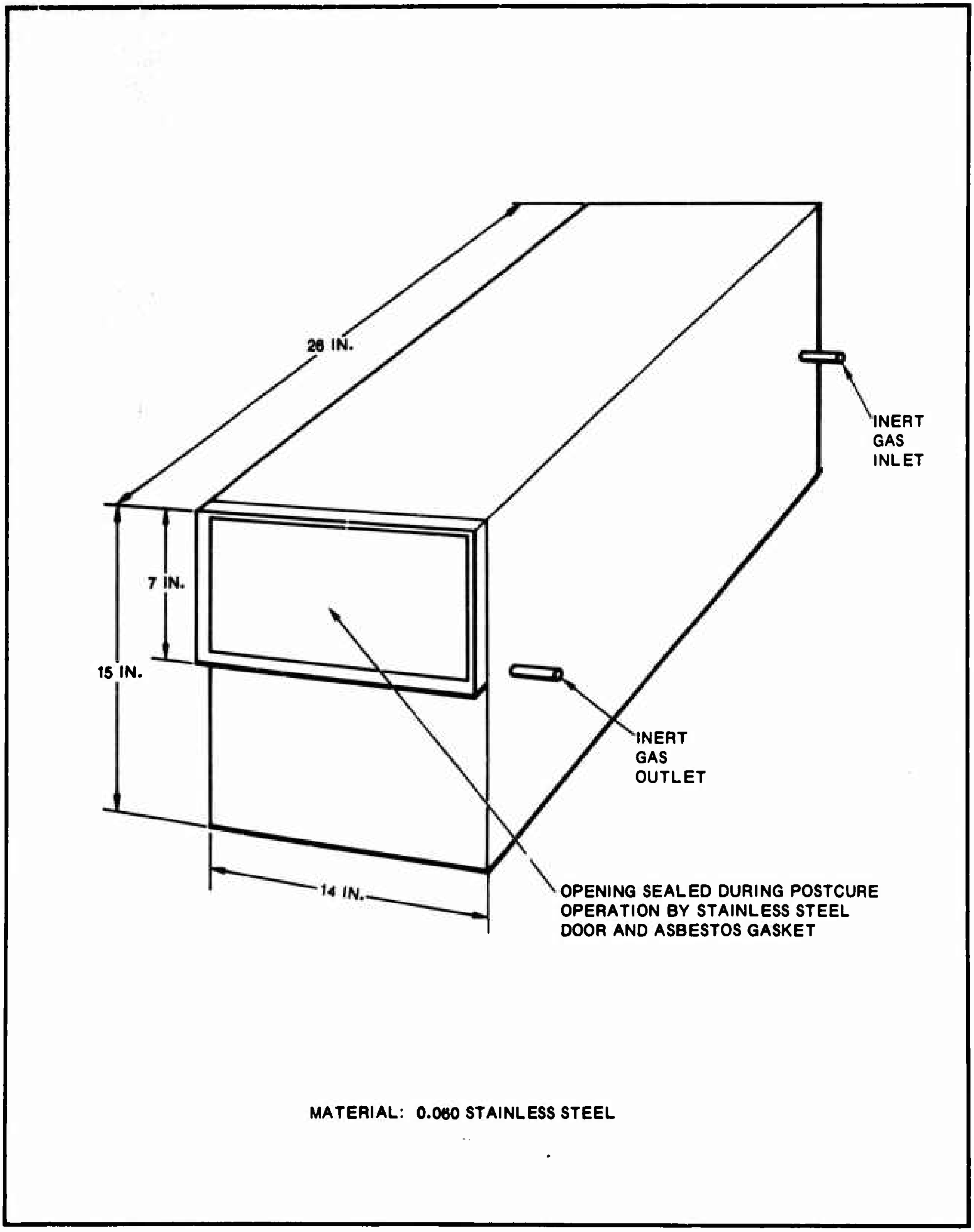



Figure 21 - Postcure Chamber for PBI Honeycomb Core Sandwich Panels

Figure 22 - Postcure Chamber Positioned in Oven

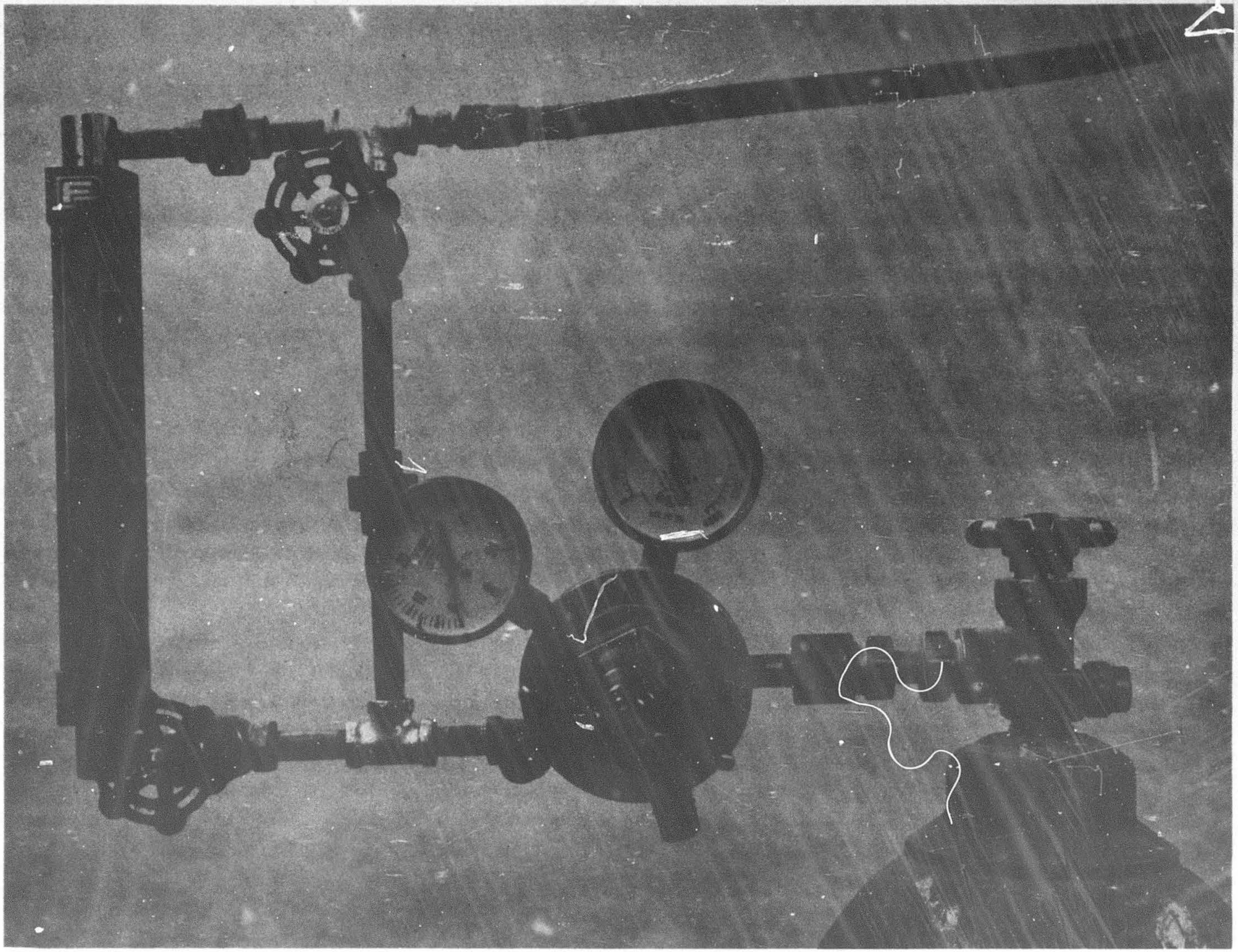


Figure 23 - Metering Device for Inert Gas Feed to Postcuring Chamber

## TABLE XXVII

### PBI(2803) Face Sheet Laminates Test Program on New Postcure Chamber

| Sample No. | Fabrication Method | Thickness | Resin Content, % | Flexural Strength PSI |
|---|---|---|---|---|
| 124 | Autoclave | .0315 | 22.4 | 68,800 |
| 125 | Autoclave | .038 | 25.3 | 42,875 |
| 126 | Autoclave | .032 | 21.3 | 43,767 |
| 127 | Press | .037 | 25.4 | 43,933 |
| 128 | Press | .033 | 22.2 | 51,800 |

Note:

1. All panels were postcured under nitrogen in the new postcure chamber using the standard PBI postcure schedule.

## TABLE XXVIII

### Controlled Postcure Test PBI(2803) Laminate

| Condition | Resin Content % | Flexural Strength PSI |
|---|---|---|
| No Postcure | 23.5 | 40,800 |
| Postcured in Air | 15.6 | 10,347 |
| Postcured in s/s envelope - $N_2$ | 23.4 | 89,900 |
| Postcured in new chamber - $N_2$ | 23.1 | 61,270 |

Notes:

1. Laminate #129 was used in this test.
2. Laminate #129 was prepared by single-stage autoclave cure using the pressure point cure method.

The results of the final evaluation tests are shown in Table XXIX. Based on the results shown in Table XXIX, argon was used as the inert gas blanket for all post curing operations conducted on the PPI (2803) sandwich composites fabricated for the Phase II study.

## TABLE XXIX

**Postcure Study on PBI(2803) Laminates**

| | SAMPLE NUMBER | | | | | |
|---|---|---|---|---|---|---|
| | 130 | | 131 | | 132 | |
| CONDITION | Resin Content % | Flexural Strength PSI | Resin Content % | Flexural Strength PSI | Resin Content % | Flexural Strength PSI |
| No Postcure | 33.0 | 52,900 | 34.1 | 60,700 | 25.0 | 28,325 |
| Postcured in Air | 27.2 | 25,000 | 29.9 | 37,570 | | |
| Postcured under nitrogen | 34.0 | 51,900 | 35.1 | 64,070 | 24.5 | 59,500 |
| Postcured under argon | | | | | 25.0 | 81,275 |

**NOTES:**

1. Laminates were prepared by single stage autoclave cure using the pressure point cure method.

2. Commercially pure nitrogen and argon were used.

# SECTION VII

## HEAT AGING STUDY - POLYIMIDE COMPOSITES

### A. GENERAL

Based on the results of the processing study (described in detail in Section IV), it was decided that the polyimide sandwich composites used in the heat aging study should be fabricated by the single-stage cure technique. The processing for the single-stage cure of the polyimide honeycomb core sandwich test panels is outlined in Section IV-D.

The test program consisted of:

Flatwise tensile strength
Compressive strength
Shear and Modulus - Ribbon Direction
Shear and Modulus - Transverse Direction

The heat aging schedule was:

Room temp. - No aging
400F - No aging, 300 hrs, 600 hrs.
500F - No aging, 100, 300, 500, 600 hrs.
600F - No aging, 100, 300, 500, 600 hrs.

All tests were performed at the aging temperature.

At the start of the program it was specified that, if a 50% reduction in shear strength in the ribbon direction had not been recorded after 600 hours at any temperature, then additional samples would be tested for shear and modulus in the ribbon direction at 200-hour intervals until a 50% reduction in shear strength had been noted.

As the heat aging program progressed it became apparent that the heat resistance of the polyimide resin system was so great that all sandwich configurations would still be exhibiting satisfactory strength values at the time the project terminated. Polyimide heat aging tests - while run as long as possible - were nevertheless discontinued with ribbon shear and modulus values still above 50% of room temperature strength.

### B. QUALITY ASSURANCE

Figure 24 shows a stack of polyimide sandwich panels prepared by the single stage cure method. Before a panel was acceptable for heat-age testing, it was checked carefully for quality in the following manner.

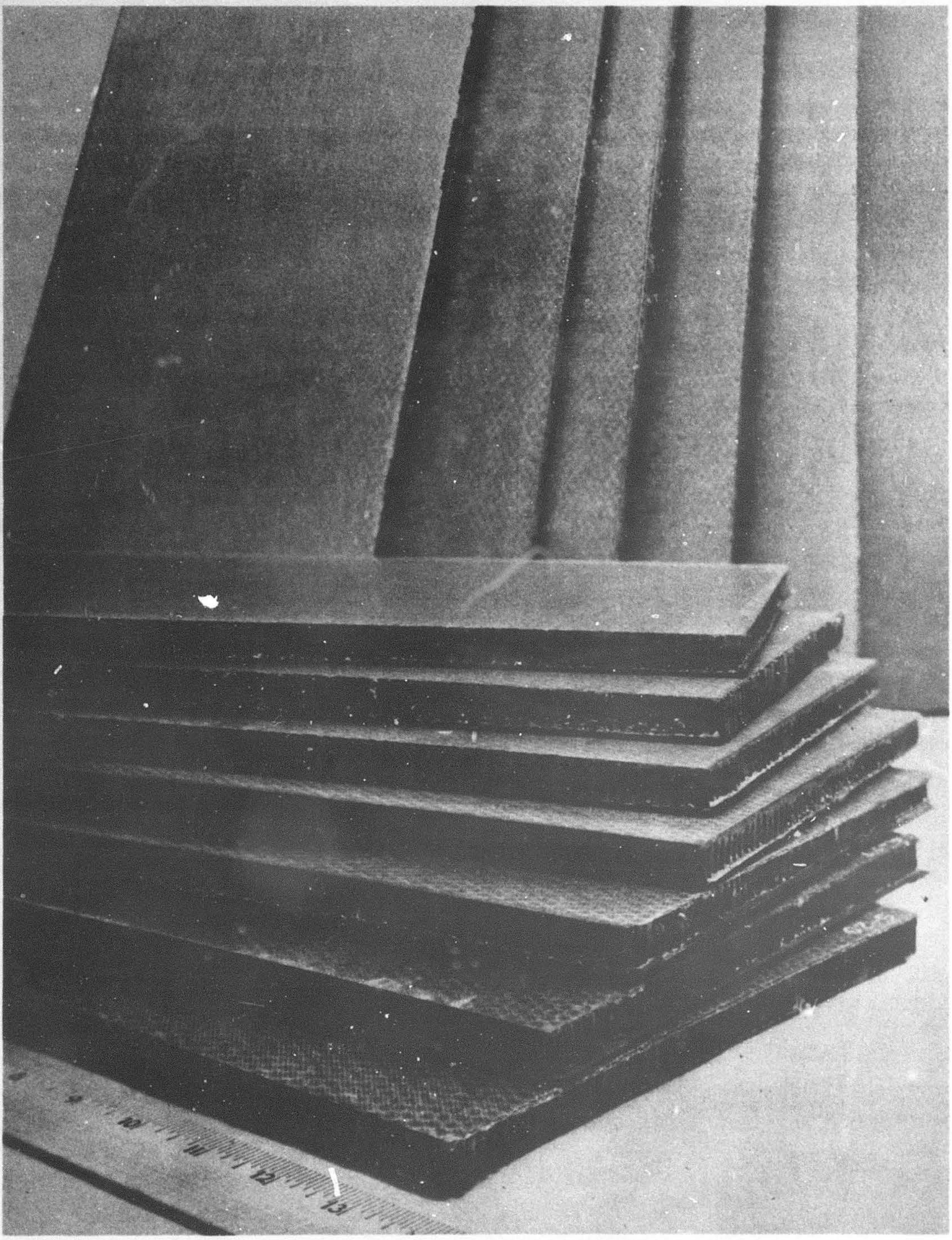


Figure 24 - Polyimide Sandwich Composites

1. Visual observation.

2. Bond testing by means of the "coin tapping" technique over the entire surface of both face sheets.

3. If the panel passed the visual and tap tests, it was cut into test specimens according to the configuration shown in Figure 25. The indicated "Quality Assurance" control specimens were then tested:

   a. Two flexural specimens in the ribbon direction.

   b. One flexural specimen in the transverse direction.

   c. Two compressive specimens 1" × 1".

   d. Three flatwise tensile specimens 1" × 1".

The Quality Assurance test results were the ultimate criteria for acceptance of a panel for the heat aging program.

Table XXX presents the results of the quality assurance tests on the 4.0 lb. density polyimide sandwich composites. Table XXXI presents the results of quality assurance tests on the 8.0 lb. density composites.

## C. HEAT AGING

The test specimens were initially cut oversize and heat aged in that condition. After the aging period, the specimens were machined to exact size just prior to testing. This practice was followed to reduce to a minimum the excessive edge effect that could occur from surface oxidation.

The heat aging specimens were stacked on special support carriers (Figure 26) during the aging period. The special support units permitted air to flow around each test specimen and assured uniform heat aging.

Heat aging was done in circulating hot air electric ovens. Oven accuracy was ±5F at the aging temperature. Figures 27, 28, and 29 show the ovens used for the heat aging program.

An outline of the specified heat aging schedule follows:

1. Room temperature

   a. No aging

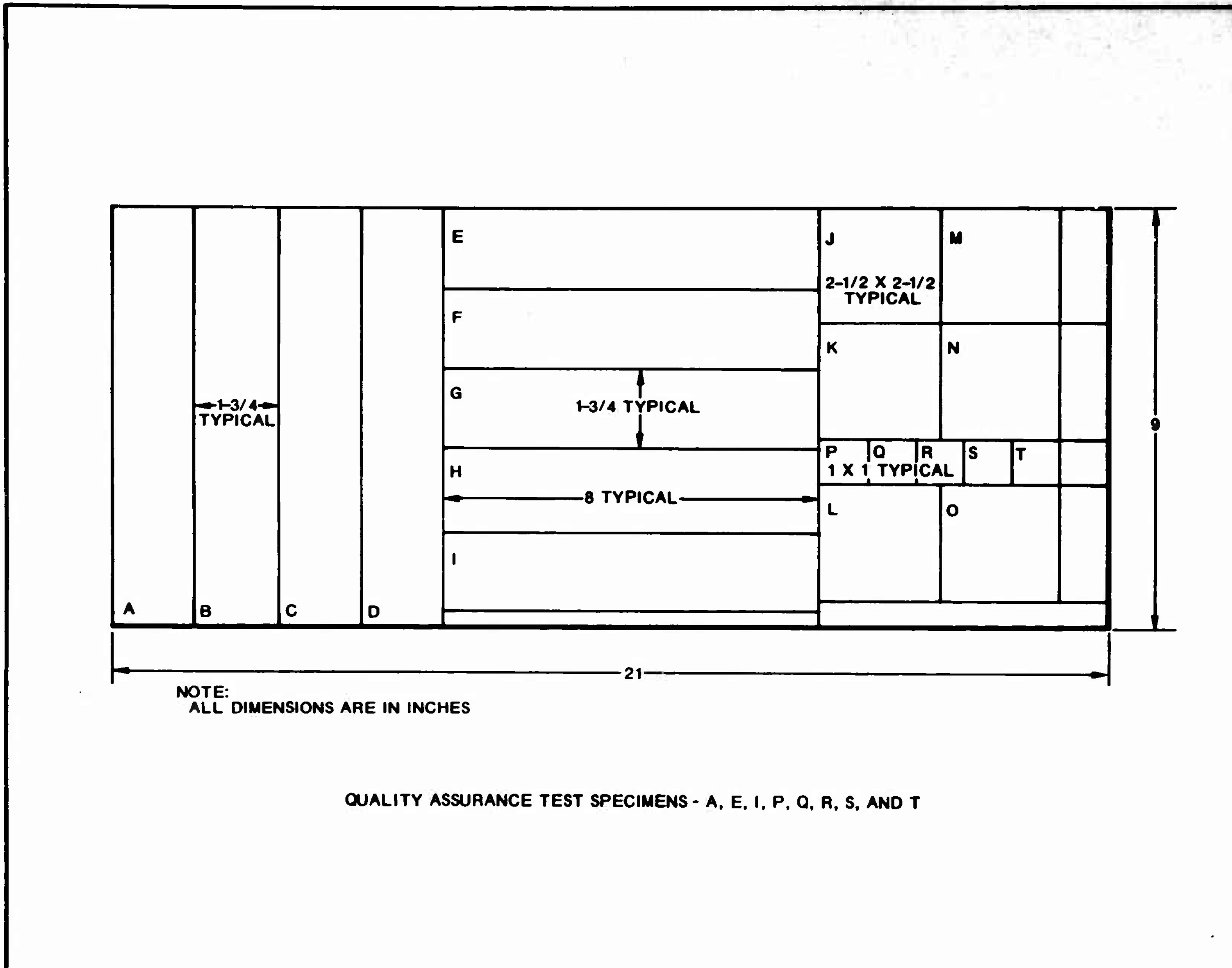



Figure 25 - Pattern for Polyimide Heat Aging Test Panels

## TABLE XXX

**Polyimide Sandwich Panels - 4.0 lb. Density Core**
**Quality Assurance Test Results**

| Panel No. | Test Condition | Flatwise Tensile Strength | Compressive Strength | Flexural Shear Strength (Ribbon Direction) | Composite Bending Modulus (Ribbon Direction) | Flexural Shear Strength (Transverse Direction) | Composite Bending Modulus (Transverse Direction) |
|---|---|---|---|---|---|---|---|
| 51 | R. T. Q. C. * | 701 | 892.5 | 442 | 4310 | 205 | 3570 |
| 52 | " | 645 | 672.5 | 424 | 4170 | 207 | 3295 |
| 54 | " | 643 | 637.5 | 373 | 4420 | 198 | 3510 |
| 55 | " | 680 | 427.5 | 352 | 4340 | 159 | 3065 |
| 50 | " | 591 | 672 | 402 | 4150 | 166 | 3080 |
| 56 | " | 679 | 535 | 346 | 4390 | 158 | 3520 |
| 44 | " | 603 | 945 | 418 | 4370 | 210 | 3460 |
| 45 | " | 548 | 677 | 428 | 4230 | 184 | 3215 |
| 47 | " | 465 | 1295 | 548 | 4150 | 256 | 3505 |
| 20 | " | 634 | 622 | 444 | - | 297 | - |
| 42 | " | 667 | 840 | 411 | 4030 | 211 | 3265 |
| 22 | " | 596 | 845 | 441 | - | 265 | - |
| 23 | " | 628 | 545 | 310 | - | 146 | - |
| 41 | " | 529 | 1120 | 429 | 4310 | 252 | 3500 |

*Quality Control

## TABLE XXXI

**Polyimide Sandwich Panels - 8.0 lb. Density Core**
**Quality Assurance Test Results**

| Panel No. | Test Condition | Flatwise Tensile Strength | Compressive Strength | Flexural Shear Strength (Ribbon Direction) | Composite Bending Modulus (Ribbon Direction) | Flexural Shear Strength (Transverse Direction) | Composite Bending Modulus (Transverse Direction) |
|---|---|---|---|---|---|---|---|
| 57 | R. T. Q. C.* | 785 | 1770 | 618 | 4973 | 514 | 4373 |
| 25 | " | 730 | 1642 | 614 | - | 398 | - |
| 58 | " | 734 | 2325 | 664 | 4720 | 527 | 4347 |
| 59 | " | 828 | 1895 | 597 | 4573 | 528 | 4300 |
| 26 | " | 700 | 1685 | 640 | - | 408 | - |
| 27 | " | 810 | 2041 | 674 | - | 453 | - |
| 28 | " | 759 | 1515 | 620 | - | 385 | - |
| 31 | " | 642 | 2200 | 680 | - | 493 | - |
| 32 | " | 815 | 2005 | 713 | - | 493 | - |
| 33 | " | 708 | 1965 | 653 | - | 508 | - |
| 34 | " | 633 | 1987 | 656 | - | 521 | - |
| 36 | " | 749 | 1650 | 690 | - | 442 | - |
| 37 | " | 697 | 2390 | 686 | - | 481 | - |
| 38 | " | 686 | 2035 | 668 | - | 572 | - |

*Quality Control

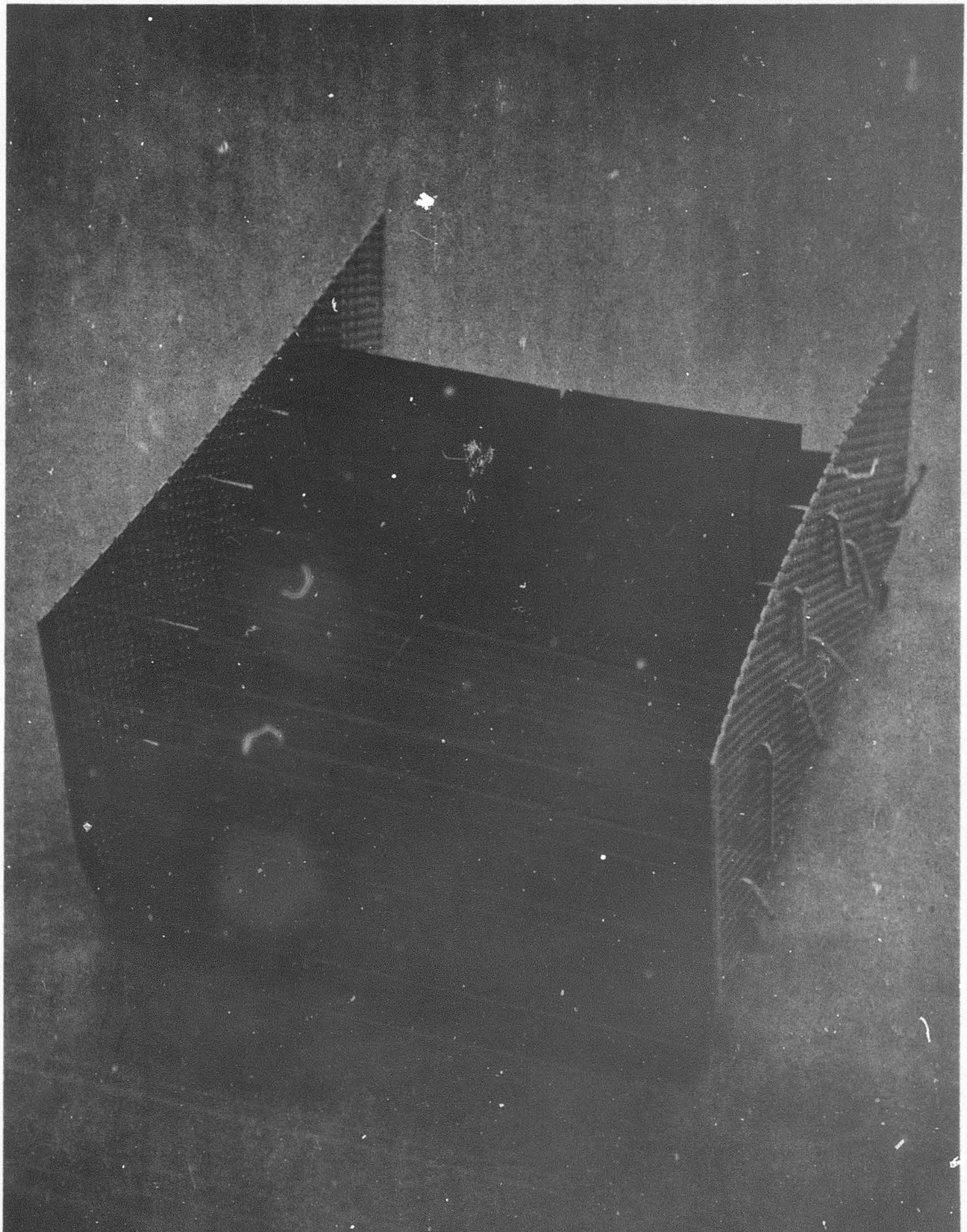


Figure 26 - Support Rack for Holding Test Specimens During Heat Aging

Figure 27 - Postcuring and Heat Aging Oven

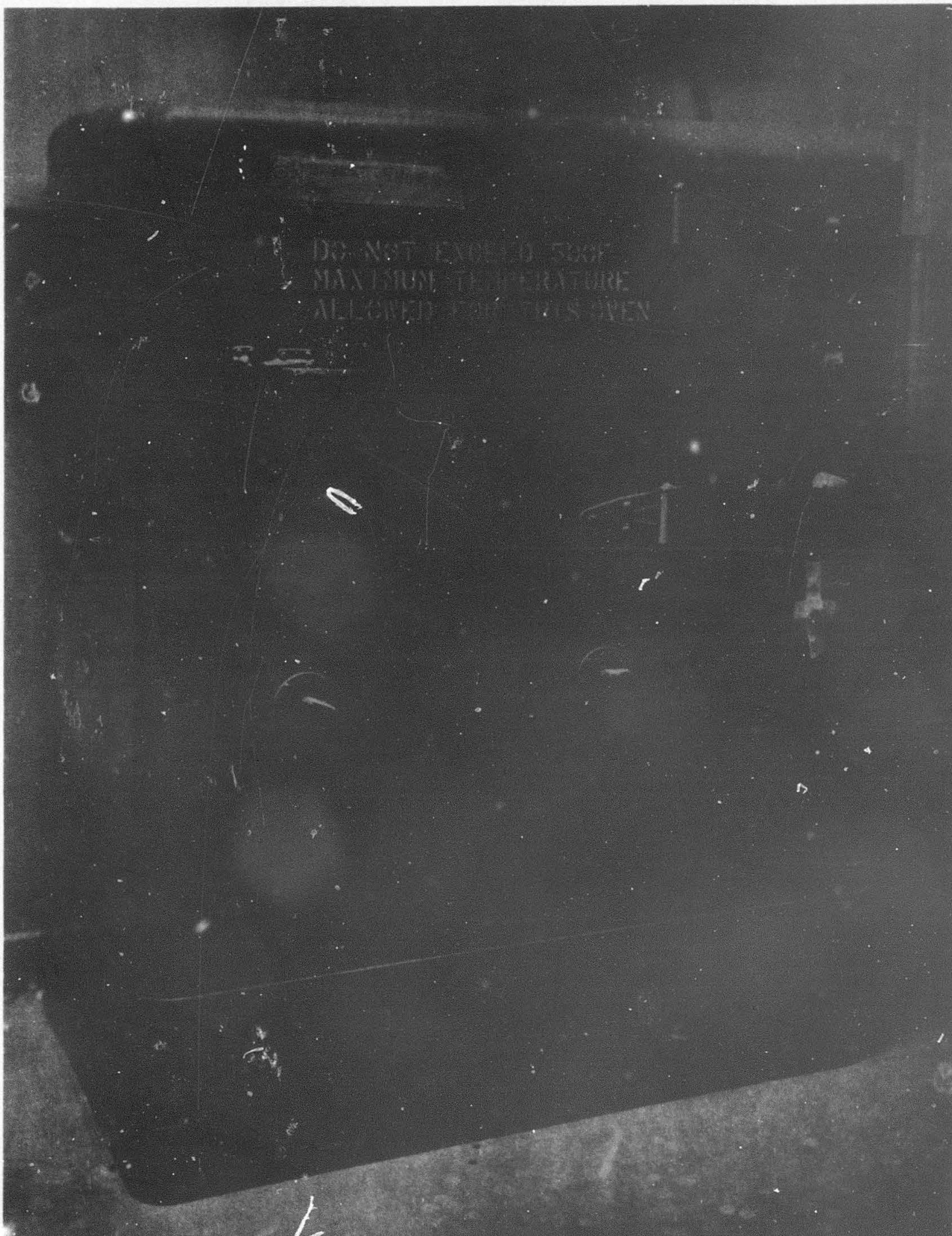


Figure 28 - Heat Aging Oven No. 10

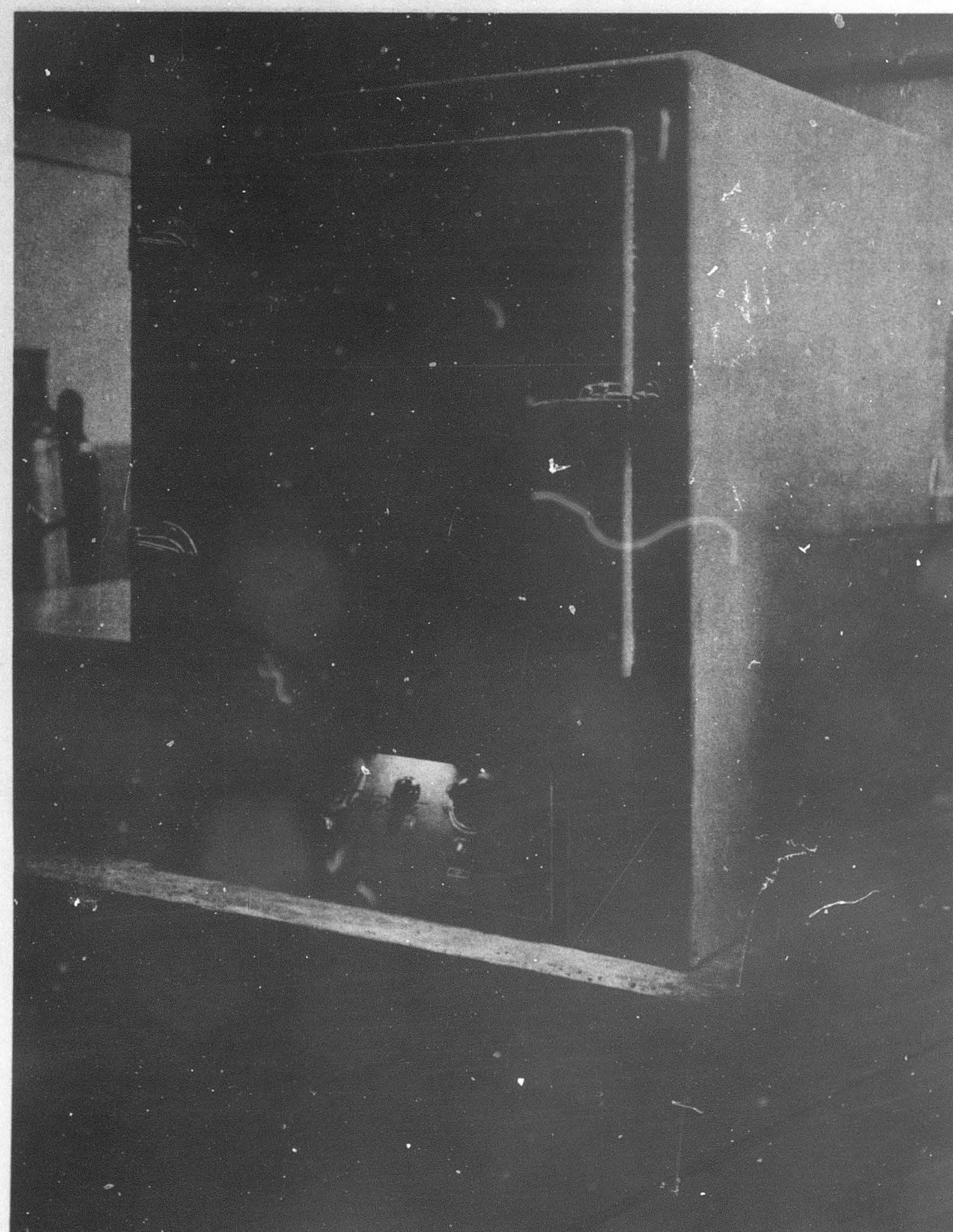


Figure 29 - Heat Aging Oven No. 11

2. 400F

   a. No aging
   b. 300 hours
   c. 600 hours

3. 500F

   a. No aging
   b. 100 hours
   c. 300 hours
   d. 500 hours
   e. 600 hours

4. 600F

   a. No aging
   b. 100 hours
   c. 300 hours
   d. 500 hours
   3. 600 hours

## D. TESTING

Testing of each set of aged specimens was completed as quickly as possible after the end of the aging period - usually within 72 hours. Testing was performed on an Instron Universal Test Machine. Figure 30 shows the GACA Instron machine with the high-temperature test chamber in position.

The test specimens were brought up to aging temperature in the Instron controlled temperature chamber. When the specimen had stabilized at the test temperature, the test was performed.

An outline of the specified testing program for the heat aged honeycomb core sandwich specimens follows:

1. Tests

   a. Flatwise tensile strength.
   b. Compressive strength.
   c. Shear strength - Ribbon Direction
   d. Shear Modulus - Ribbon Direction
   e. Shear Strength - Transverse Direction
   f. Shear Modulus - Transverse Direction

2. All tests will be made at aging temperatures.

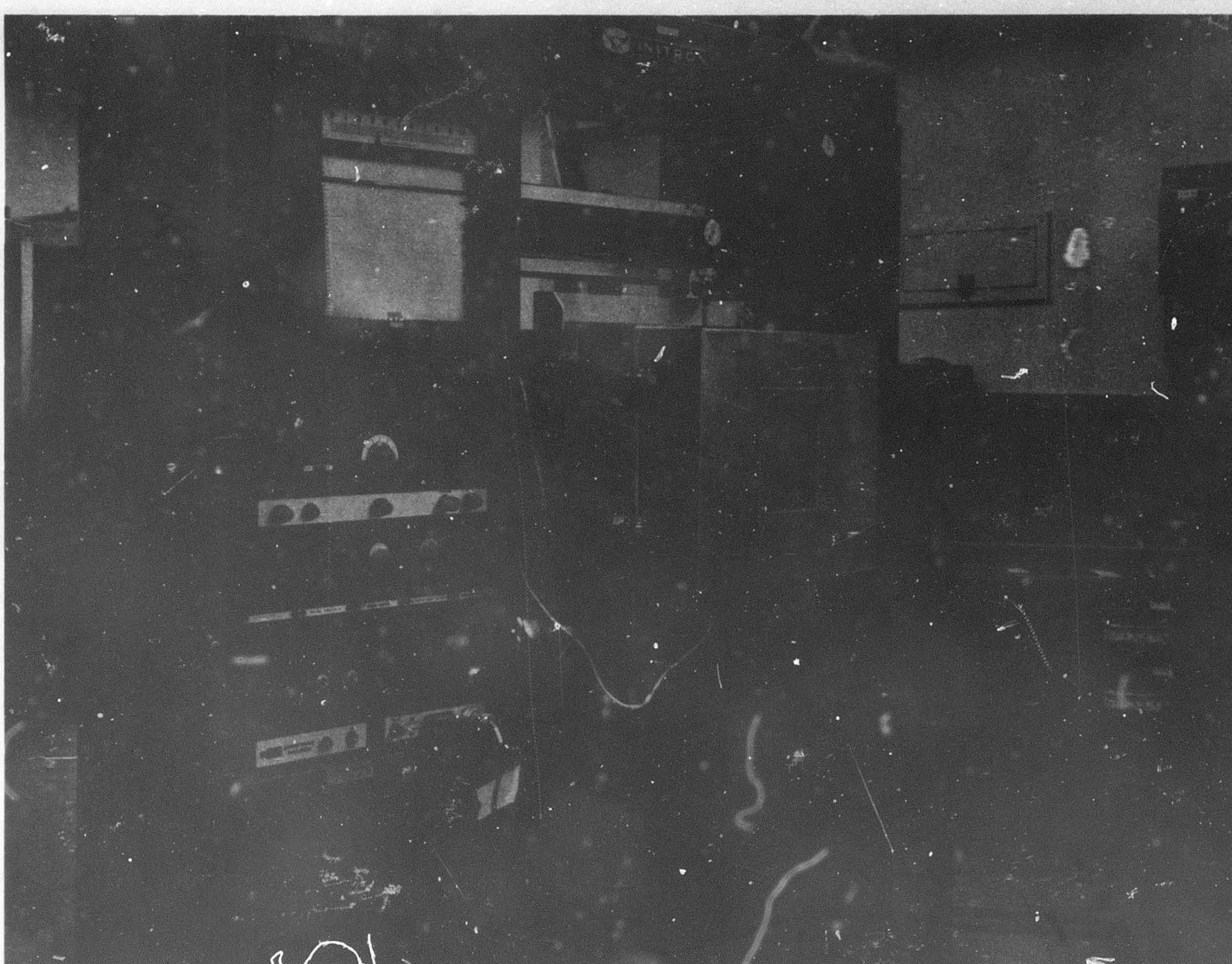


Figure 30 - Goodyear Aerospace Instron Machine

3. Length of tests will be continued until a 50% reduction in shear strength is noted.

The tests were run according to MIL-A-25463 and MIL-STD-401A specifications.

1. Flatwise Tensile Strength

This test was performed according to MIL-A-25463. Figure 31 shows a flatwise tensile specimen being tested at room temperature.

Flatwise tensile data provided an excellent study on the heat resistant capabilities of the face sheet to core adhesive bond.

The only problem initially anticipated with this test was the adequacy of the bond adhering the tensile loading blocks to the face sheets at the test temperatures of 500F and 600F.

A modified polyimide adhesive was employed in this program to bond the loading blocks to the sample for the 500F and 600F tests.

For the room temperature and 400F tests, a modified low temperature curing epoxy adhesive was used to bond the loading blocks to the flatwise tensile specimens.

2. Compressive Strength

This test was performed according to MIL-STD-401A. The compressive strength test provided excellent data from which the heat resistant properties of the honeycomb core could be determined.

Figure 32 shows a compressive sample in position for testing in the high-temperature chamber.

3. Shear Strength

Flexural shear strengths were determined according to MIL-A-25463. The flexural test was considered an excellent test because it approximated the actual conditions of service for sandwich constructions and was simple to perform.

Core shear values for the honeycomb sandwich specimens were obtained from a 1-1/4" × 8" flexural specimen. The specimen was simply supported on a 6" span and loading was applied at two quarter span points.

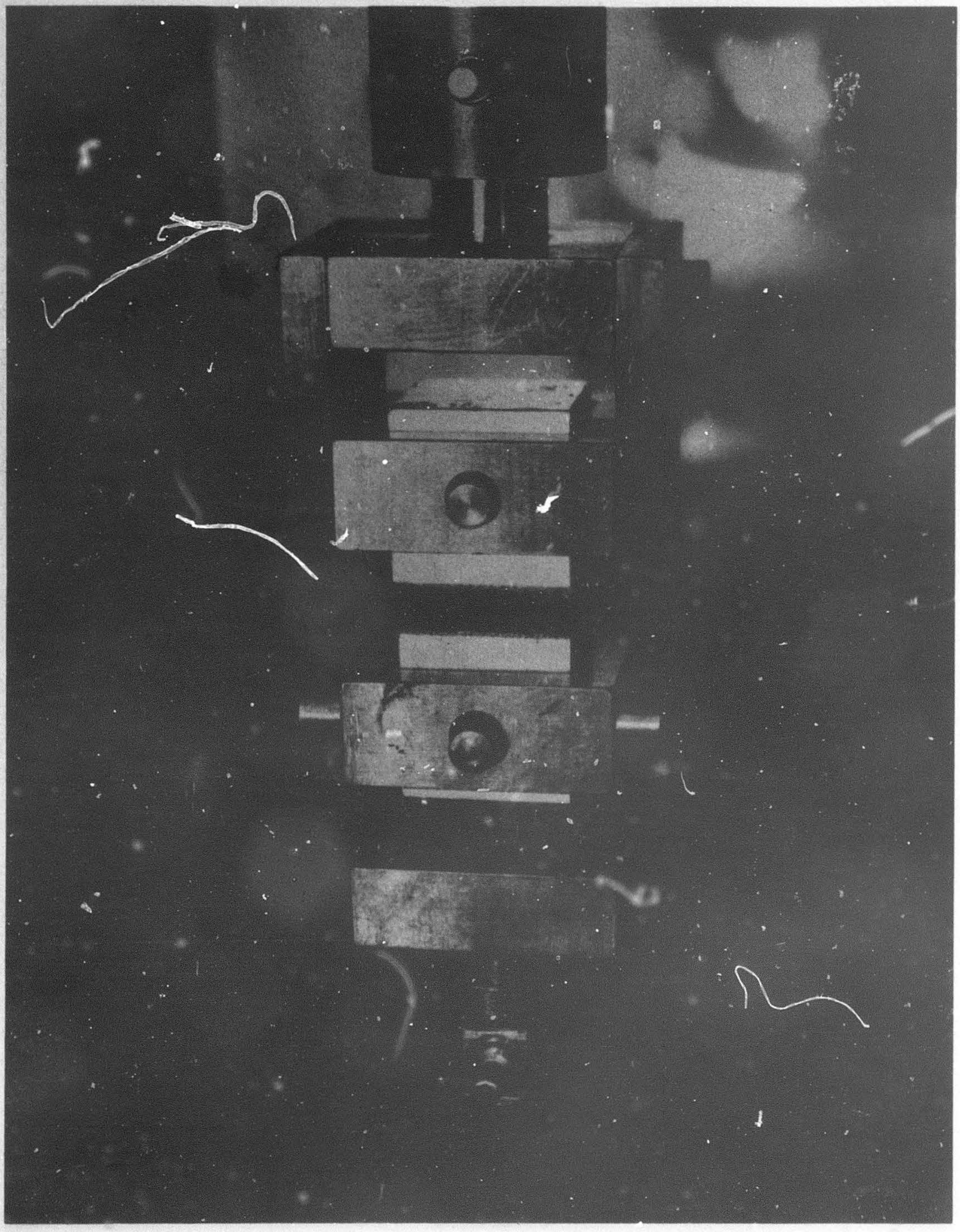


Figure 31 - Flatwise Tensile Specimens Being Tested in the Instron Instrument

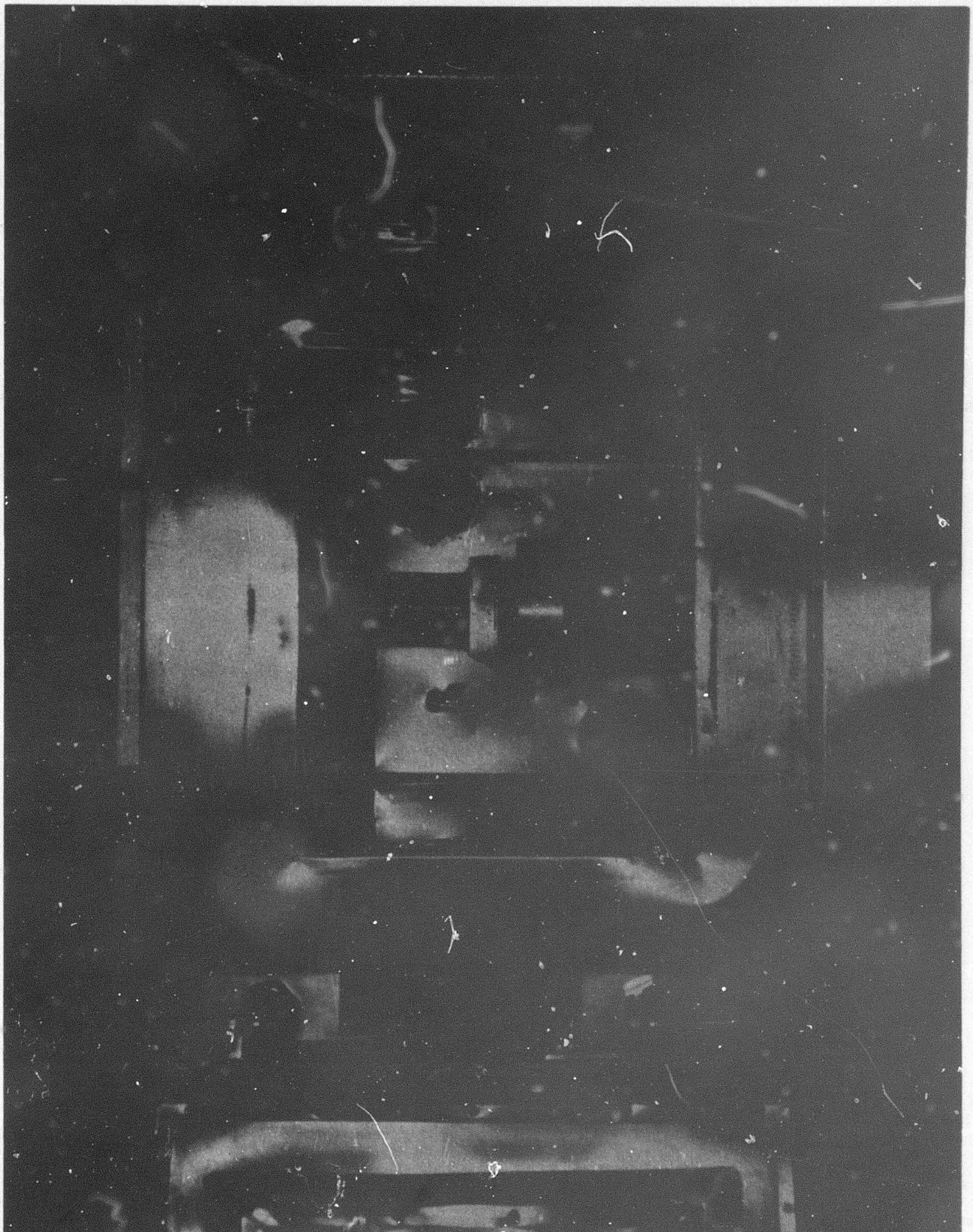


Figure 32 - Testing Compressive Sample in High-Temperature Chamber

The specimen configuration was ideal for developing core shear values for this project. The distance between the support and load spans was sufficiently short to insure that failure would occur by core shear rupture. The specimen was large enough to diminish the effects of minor dimensional variations, but not so large as to be economically inefficient.

Figure 33 shows the interior of the Instron high-temperature chamber with a flexural specimen in position for testing. The test data obtained from the flexural tests provided an excellent indication of the performance of the overall sandwich composite.

4. Shear Modulus

Core shear modulus values for the honeycomb sandwich specimens were obtained from the 1-1/4" × 8" flexural specimens. These modulus values were a measure of beam stiffness for the sandwich configuration and loading conditions of the particular specimens. It was identified as Composite Bending Modulus, Q.

This modulus value was obtained by recording deflection during performance of the flexural shear strength test. The initial slope of the load-stroke curve determined the value of Q.

Figure 34 shows a close-up view of the high-temperature deflectomer which produced a controlled stress-strain curve for determining modulus values. Figure 35 shows a section of chart paper taken from the Instron machine. Modulus values were calculated from the stress-strain curves produced on the chart.

The Composite Bending Modulus, Q, was influenced by the bending stiffness of the face sheets, as well as the shear stiffness of the core. The Composite Bending Modulus provided a logical descriptive value on the performance of the composite. It allowed us to prepare an excellent comparative chart of the performance of the various sandwich composite configurations through the heat aging environments.

## E. TEST RESULTS

Table XXXII presents the test results on the 4.0 lb density polyimide honeycomb core sandwich composites through 600 hours of heat aging. Table XXXIII presents the test results on the 8.0 lb. density polyimide composites through the 600 hour period.

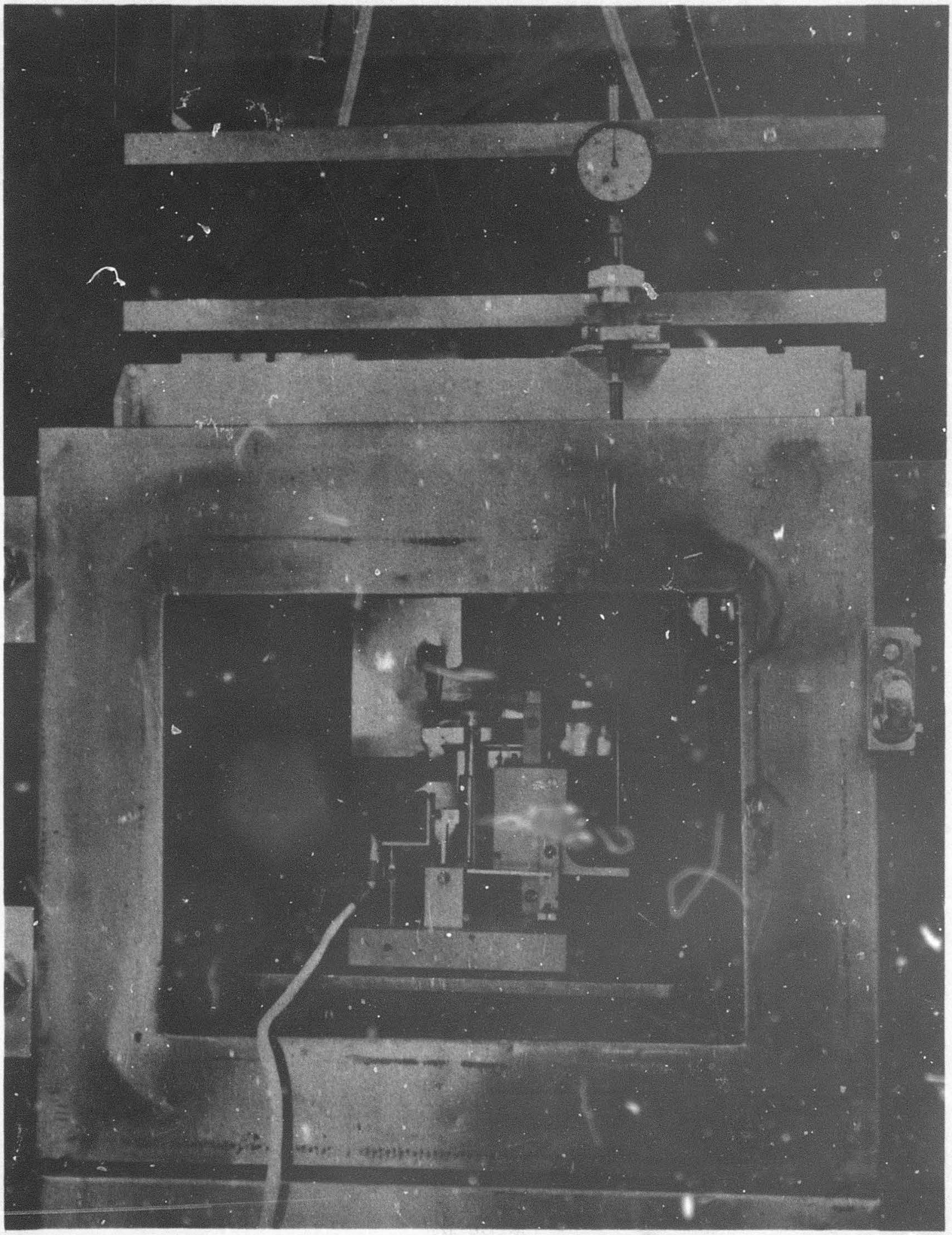


Figure 33 - Flexural Specimen in Position for High-Temperature Testing

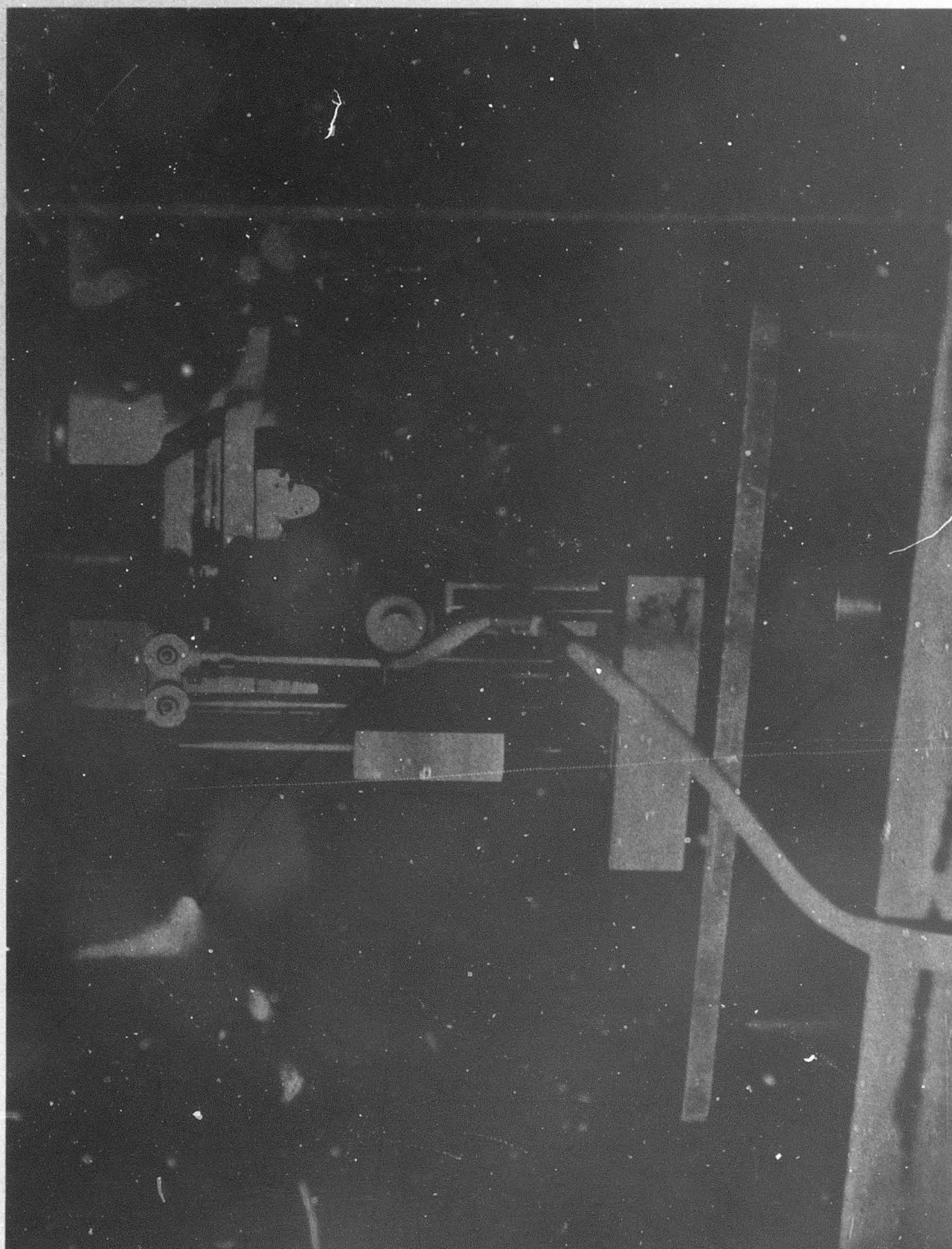


Figure 34 - High-Temperature Deflectometer in Position

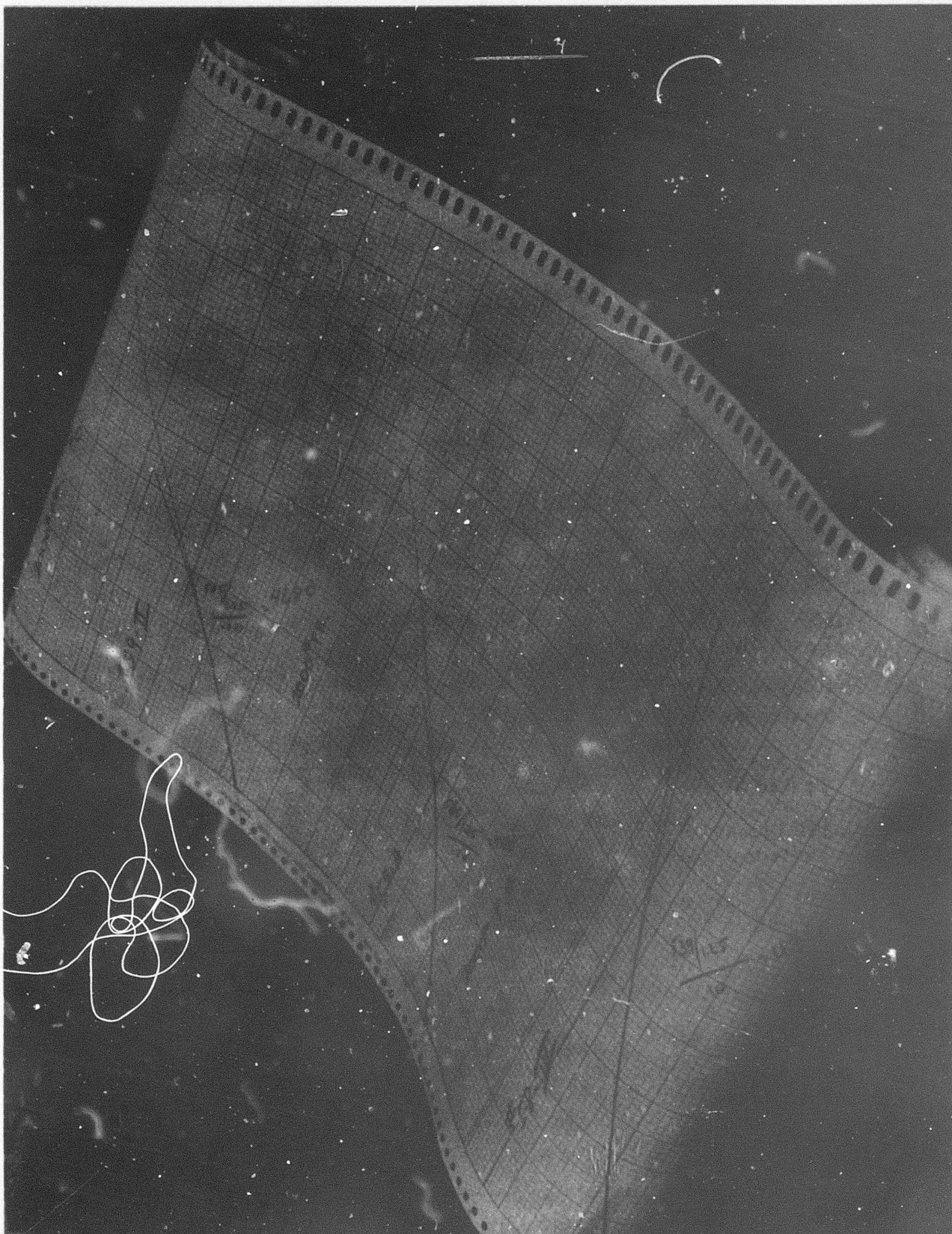


Figure 35 - Instron Chart with Stress-Strain Curves and Modulus Calculations

## TABLE XXXII

### Polyimide Sandwich Panels - 4.0 Density Core

### Heat Aging Test Results

| NO. | CONDITION | Flat-wise Ten-sile | Comp. Strength | Flex-ural "L" Shear | Com-posite Core Mod. | Flex-ural "W" Shear | Com-posite Core Mod. |
|---|---|---|---|---|---|---|---|
| 51 | No Aging Tested at R. T. | 665 | 970 | 438 | 4303 | 228 | 3500 |
| 52 | No Aging Tested at 400F | 424 | 448 | 319 | 3680 | 169 | 3030 |
| 54 | No Aging Tested at 500F | 457 | 456 | 249 | 3173 | 154 | 2217 |
| 55 | No Aging Tested at 600F | 303 | 158 | 151 | 3650 | 125 | 1423 |
| 50 | 100 hrs at 500F Tested at 500F | 334 | 471 | 319 | 3680 | 169 | 3030 |
| 56 | 100 hrs at 600F Tested at 600F | 413 | 289 | 236 | 2220 | 134 | 1227 |
| 44 | 300 hrs at 400F Tested at 400F | 488 | 469 | 355 | 3893 | 181 | 2993 |
| 45 | 300 hrs at 500F Tested at 500F | 421 | 473 | 290 | 4083 | 166 | 3163 |
| 47 | 300 hrs at 600F Tested at 600F | 363 | 575 | 295 | 2887 | 167 | 1943 |
| 20 | 500 hrs at 500F Tested at 500F | 345 | 420 | 277 | 4153 | 196 | 2817 |
| 42 | 500 hrs at 600F Tested at 600F | 352 | 308 | 243 | 5927 | 125 | 5275 |
| 22 | 600 hrs at 400F Tested at 400F | 398 | 665 | 323 | 4433 | 196 | 2587 |
| 23 | 600 hrs at 500F Tested at 500F | 449 | 263 | 204 | 3397 | 116 | 1983 |
| 41 | 600 hrs at 600F Tested at 600F | 360 | 303 | 229 | 3073 | 144 | 2087 |

## TABLE XXXIII

### Polyimide Sandwich Panels - 8.0 Density Core

### Heat Aging Test Results

| NO. | CONDITION | Flat-wise Ten-sile | Comp. Strength | Flex-ural "L" Shear | Com-posite Core Mod | Flex-ural "W" Shear | Com-posite Core Mod |
|---|---|---|---|---|---|---|---|
| 57 | No Aging Tested at R. T. | 818 | 1799 | 627 | 4583 | 513 | 4397 |
| 25 | No Aging Tested at 400F | 537 | 874 | 356 | 2717 | 309 | 1640 |
| 58 | No Aging Tested at 500F | 544 | 997 | 312 | 3300 | 300 | 3073 |
| 59 | No Aging Tested at 600F | 416 | 260 | 225 | 4520 | 174 | 1697 |
| 26 | 100 hrs at 500F Tested at 500F | 452 | 631 | 305 | 5327 | 244 | 3500 |
| 27 | 100 hrs at 600F Tested at 600F | 362 | 907 | 261 | 3600 | 224 | 2267 |
| 28 | 300 hrs at 400F Tested at 400F | 517 | 768 | 345 | 2355 | 267 | 1847 |
| 31 | 300 hrs at 500F Tested at 500F | 456 | 911 | 357 | 4853 | 232 | 3613 |
| 32 | 300 hrs at 600F Tested at 600F | 392 | 490 | 290 | 3603 | 229 | 3350 |
| 33 | 500 hrs at 500F Tested at 500F | 412 | 752 | 354 | 4050 | 273 | 3697 |
| 34 | 500 hrs at 600F Tested at 600F | 293 | 499 | 316 | 3630 | 237 | 2790 |
| 36 | 600 hrs at 400F Tested at 400F | 553 | 868 | 407 | 5773 | 303 | 4270 |
| 37 | 600 hrs at 500F Tested at 500F | 368 | 923 | 390 | 4597 | 297 | 3335 |
| 38 | 600 hrs at 600F Tested at 600F | 383 | 475 | 281 | 3440 | 226 | 2640 |
| 24 | 800 hrs at 600F Tested at 500F | 383 | 333 | 276 | 4637 | 198 | 3253 |

It was noted with interest that the strength values for the polyimide sandwich composites remained high at all temperature levels through the 600 hour aging period. The flexural shear strengths (ribbon direction) of both the 4.0 lb. and the 8.0 lb. density core samples were above 50% of the room temperature value after the 600 hour heat aging period at 400F, 500F, and 600F. (The one exception to this was the 8.0 lb. density core sample aged at 600F. The shear strength was only 42% of the room temperature value after 600 hours aging, but appeared to be on an increasing slope).

In view of these encouraging results, it was decided to continue the heat aging tests. Shear strength and modulus in the ribbon direction were determined at 200 hour intervals beyond the 600 hour periods according to the schedule shown below.

1. 4.0 lb. density core composite.

   a. 400F aging temperature - a total of 1400 hours
   b. 500F aging temperature - a total of 1400 hours
   c. 600F aging temperature - a total of 1000 hours

2. 8.0 lb. density core composite

   a. 400F aging temperature - a total of 1400 hours
   b. 500F aging temperature - a total of 1200 hours
   c. 600F aging temperature - a total of 800 hours

Table XXXIV presents the results of the additional heat aging tests performed on the 4.0 lb. core density polyimide sandwich composites. Table XXXV presents the results of the additional heat aging tests performed on the 8.0 lb. core density polyimide sandwich composites.

It should be noted that time did not permit obtaining all additional heat aging information originally scheduled.

## TABLE XXXIV

### Polyimide Sandwich Panels - 4.0 Density Core
### Heat Aging Test Results

| No. | Condition | Flexural Ribbon Shear | Composite Core Modulus |
|---|---|---|---|
| 69A | Control | 370 | 4470 |
| 69B | 1000 hrs at 400F<br>Tested at 400F | 304 | 3123 |
| 69C | 1200 hrs at 400F<br>Tested at 400F | 263 | 3313 |
| 69D | 1400 hrs at 400F<br>Tested at 400F | 254 | 3000 |
| 64A | Control | 361 | 4063 |
| 64B | 800 hrs at 500F<br>Tested at 500F | 241 | 3203 |
| 64C | 1000 hrs at 500F<br>Tested at 500F | 253 | 3543 |
| 64D | 1200 hrs at 500F<br>Tested at 500F | 228 | 2870 |
| 64E | 1400 hrs at 500F<br>Tested at 500F | 211 | 3163 |

## TABLE XXXV

### Polyimide Sandwich Panels - 8.0 Density Core

### Heat Aging Test Results

| No. | Condition | Flexural Ribbon Shear | Composite Core Modulus |
|---|---|---|---|
| 63A | Control | 615 | 4485 |
| 63B | 800 hrs at 400F<br>Tested at 400F | 439 | 4007 |
| 63C | 1000 hrs at 400F<br>Tested at 400F | 405 | 3740 |
| 63D | 1200 hrs at 400F<br>Tested at 400F | 398 | 3780 |
| 63E | 1400 hrs at 400F<br>Tested at 400F | 393 | 3650 |
| 70A | Control | 494 | 4270 |
| 70B | 800 hrs at 500F<br>Tested at 500F | 326 | 3790 |
| 70C | 1000 hrs at 500F<br>Tested at 500F | 297 | 3820 |

## SECTION VIII

## HEAT AGING STUDY - PBI SANDWICH COMPOSITE

### A. GENERAL

Based on the results of the processing study discussed in Section VI the single-stage cure technique using the pressure-point method was selected for preparing the PBI (2803) sandwich composites to be evaluated in the Phase II heat aging study.

The heat aging schedule was the same as for the polyimide composites.

Room temperature - No aging
400F - No aging, 300 hours, 600 hours
500F - No aging, 100, 300, 500, 600 hours
600F - No aging, 100, 300, 500, 600 hours

The test program was also the same:

Flatwise tensile strength
Compressive Strength
Shear and Modulus - Ribbon Direction
Shear and Modulus - Transverse Direction

All tests were performed at the aging temperature.

At the start of the program it was specified that those series which maintained shear strength values greater than 50% of their room temperature values after 600 hour aging should be continued. Additional samples were to be tested for shear and modulus (ribbon direction) at 200 hour intervals until a 50% reduction in shear strength occurred.

The PBI test series aged at 400F were the only ones which maintained ribbon direction shear strength values above 50% after 600 hours aging. However, time did not permit the continuation of the aging program. All PBI aging tests were terminated after the 600 hour period.

### B. QUALITY ASSURANCE

Figure 36 shows a stack of PBI (2803) sandwich panels prepared by the single stage pressure poi method. Post curing was performed under an argon atmosphere. ach panel was checked carefully for quality in the following manner before acceptance for the Phase II heat aging study.

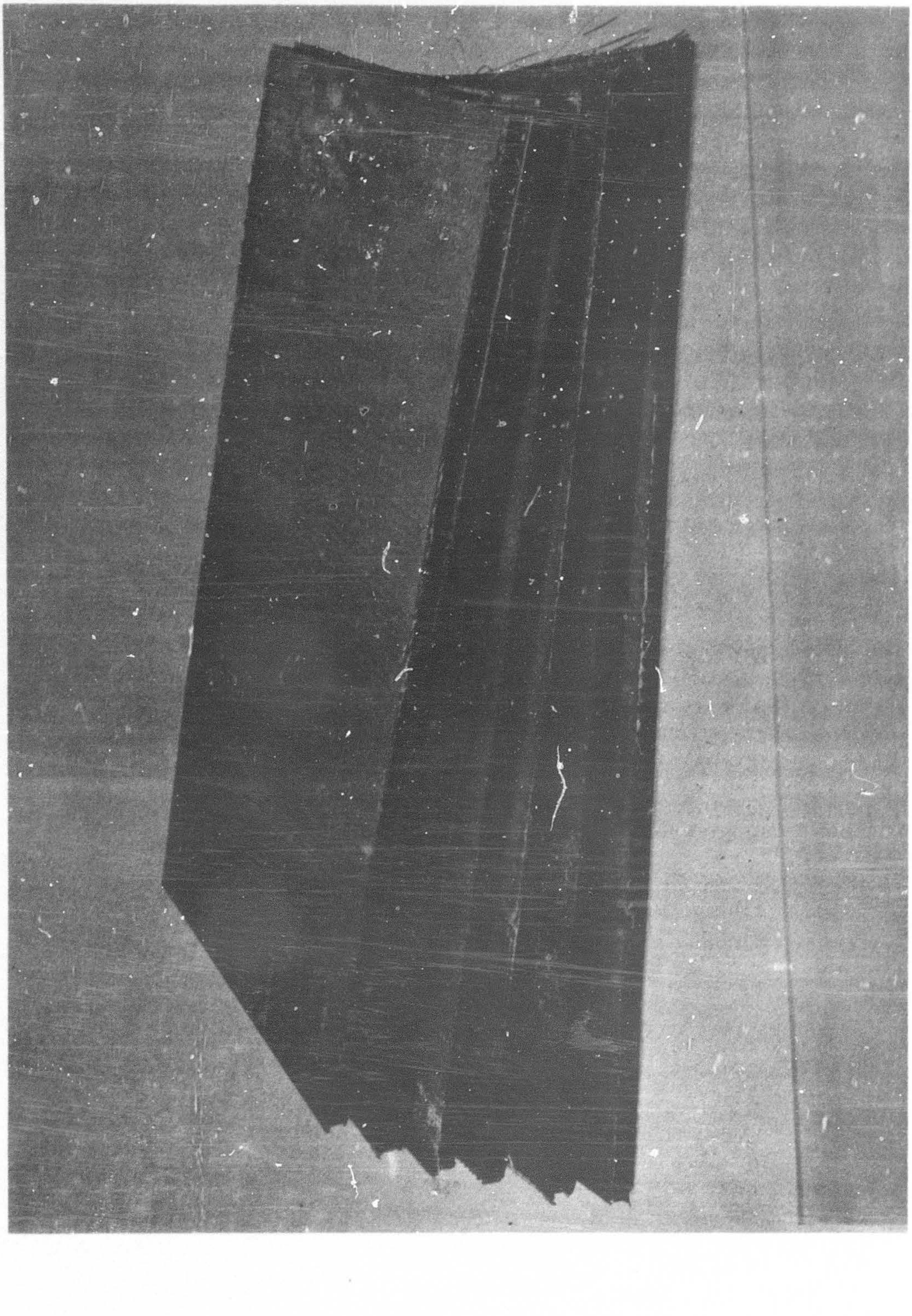


Figure 36 - PBI Sandwich Composites

1. Visual observation.

2. Bond testing by means of the "coin tapping" technique over the entire surface of both face sheets.

3. Physically testing selected control specimens from each panel.

   a. Three flexural specimens in the ribbon direction.
   b. Three flexural specimens in the transverse direction.
   c. Three compressive specimens 1" × 1".
   d. Three flatwise tensile specimens 1" × 1".

Figure 37 shows the manner in which each PBI panel was cut for heat aging and quality assurance testing. As indicated, selected test samples were obtained from each panel for Quality Assurance and Control testing. Table XXXVI presents the results of the quality assurance tests conducted on 4.0 lb. density PBI core sandwich composites. Table XXXVII presents the results of the quality assurance tests conducted on 8.0 lb. density PBI core sandwich composites.

## C. HEAT AGING

The specimens to be heat aged were initially cut oversize and then trimmed to exact size after heat aging in the hopes of reducing the edge effect of oxidation. The specimens were stacked on the special support carriers (Figure 26) during the aging period to permit free air circulation around each specimen and assure uniform heat aging.

Circulating hot air electric ovens, with an accuracy of ±5F at the aging temperature, were used for heat aging the PBI composites.

The heat aging schedule was:

1. Room temperature

   a. No aging

2. 400F

   a. No aging
   b. 300 hours
   c. 600 hours

3. 500F

   a. No aging
   b. 100 hours
   c. 300 hours

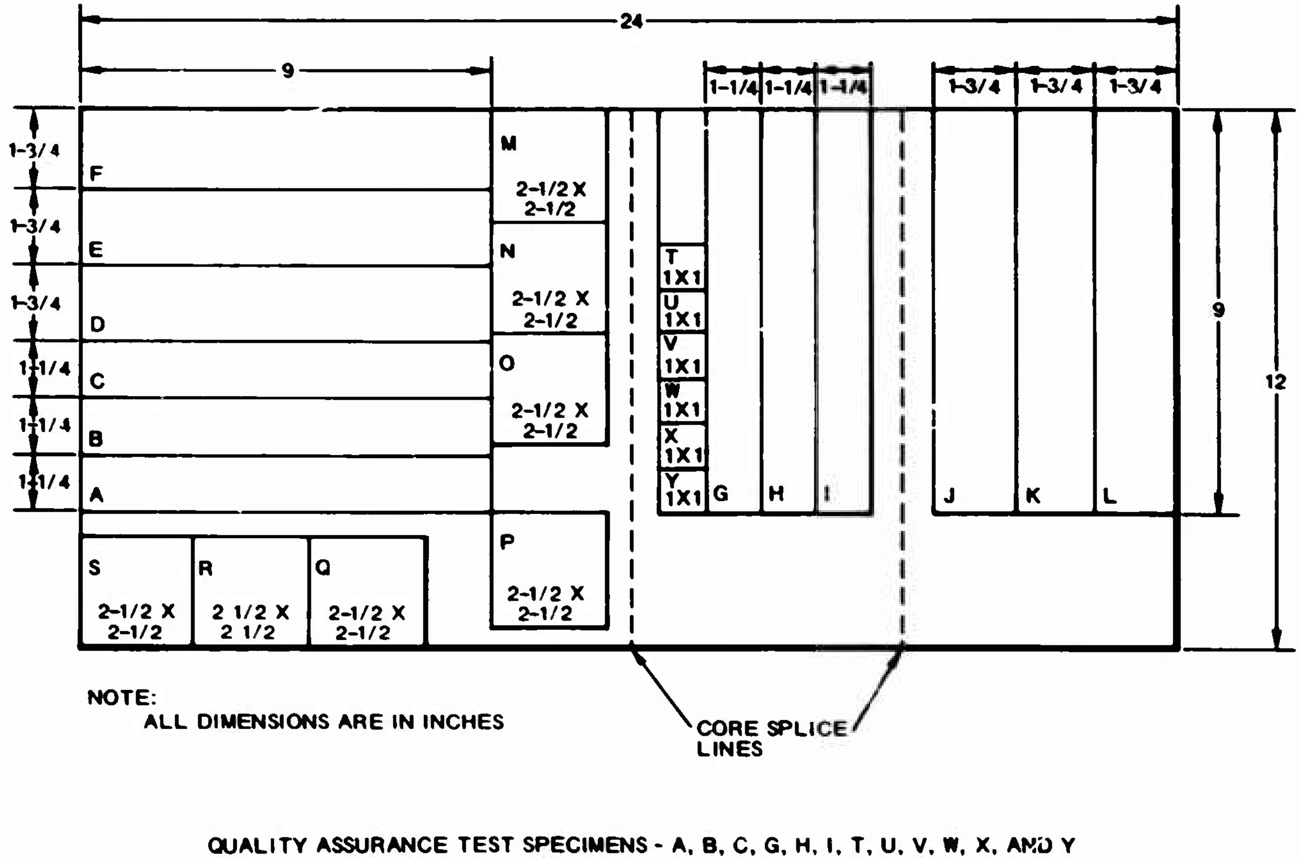



Figure 37 - Pattern for PBI Heat Aging Test Panels

## TABLE XXXVI

**PBI Sandwich Panels - 4.0 lb. Density Core**

**Quality Assurance Test Results**

| Panel No. | Test Condition | Flatwise Tensile Strength | Compressive Strength | Flexural Shear Strength (Ribbon Direction) | Composite Bending Modulus (Ribbon Direction) | Flexural Shear Strength (Transverse Direction) | Composite Bending Modulus (Transverse Direction) |
|---|---|---|---|---|---|---|---|
| 65 | R. T. Q. C.* | 331 | 1046 | - | - | 214 | 4293 |
| 90 | " | 470 | 480 | 191 | 3200 | 108 | 2417 |
| 92 | " | 274 | 382 | 198 | 3247 | 98 | 2493 |
| 93 | " | 266 | 367 | 200 | 3340 | 107 | 2657 |
| 86 | " | - | - | 210 | 3520 | 119 | 2523 |
| 88 | " | 157 | 241 | 183 | 2467 | 81 | 1455 |
| 84 | " | 243 | 1085 | 369 | 5013 | 328 | 4810 |
| 82 | " | 221 | 905 | 293 | 4803 | 261 | 4573 |
| 81 | " | 251 | 1052 | 205 | 4183 | 147 | 3950 |
| 78 | " | 295 | 1023 | 407 | 5533 | 313 | 5057 |
| 66 | " | 200 | 693 | 268 | 5003 | 177 | 3583 |
| 67 | " | 360 | 410 | 264 | 4540 | 140 | 3813 |
| 68 | " | 303 | 779 | 405 | 5103 | 222 | 4290 |
| 71 | " | 309 | 545 | 285 | 4780 | 195 | 3963 |

*Quality Control

## TABLE XXXVII

**PBI Sandwich Panels - 8.0 lb. Density Core**
**Quality Assurance Test Results**

| Panel No. | Test Condition | Flatwise Tensile Strength | Compressive Strength | Flexural Shear Strength (Ribbon Direction) | Composite Bending Modulus (Ribbon Direction) | Flexural Shear Strength (Transverse Direction) | Composite Bending Modulus (Transverse Direction) |
|---|---|---|---|---|---|---|---|
| 89 | R. T. Q. C.* | 174 | 751 | 336 | 4600 | 244 | 4395 |
| 91 | | 222 | 713 | 356 | 4665 | 263 | 4167 |
| 94 | | 209 | 929 | 417 | 4660 | 260 | 4507 |
| 96 | | 169 | 972 | 303 | 4817 | 279 | 4427 |
| 85 | | 245 | 315 | 173 | 3597 | 30 | 579 |
| 87 | | 346 | 609 | 156 | 3817 | 210 | 2090 |
| 77 | | 333 | 623 | 385 | 4710 | 142 | 3707 |
| 80 | | 294 | 34 | 208 | 4247 | 113 | 3333 |
| 97** | | 203 | 883 | 443 | 5105 | 284 | 4720 |
| 83 | | 227 | 601 | 319 | 4710 | 267 | 4063 |
| 72 | | 258 | 576 | 278 | 5277 | 284 | 4943 |
| 73 | | 300 | 652 | 367 | 5197 | 291 | 4577 |
| 74 | | 336 | 752 | 414 | 5417 | 289 | 4707 |
| 75 | | 302 | 916 | 403 | 6173 | 289 | 4997 |
| 76 | | 458 | 855 | 416 | 5830 | 255 | 4517 |

*Quality Control

** Replaced panel #80

d. 500 hours
e. 600 hours

4. 600F

a. No aging
b. 100 hours
c. 300 hours
d. 500 hours
e. 600 hours

## D. TESTING

Testing of the PBI sandwich composites was performed in the same manner as described in Section VII-D for polyimide composites.

## E. TEST RESULTS

Table XXXVIII presents the complete results of the heat aging tests performed on 4.0 lb. core density PBI sandwich composites. Table XXXIX presents the complete results of the heat aging tests performed on 8.0 lb. core density PBI sandwich composites. It should be noted that it was impossible to test any of the specimens that were heat aged at 600F. The PBI resins were so subject to oxygen degradation that hot air aging of the unprotected honeycomb core sandwich specimens, proved to be disastrous. The honeycomb core was quite porous and permeable. The skins were only .030" to .035" thick and were also permeable. Since the samples were relatively small in size, every portion of the test composite was vulnerable to oxygen attack.

Extreme care was used during fabrication to prevent oxidation.

The PBI sandwich composites were fabricated by a single stage autoclave technique. The sandwich lay-up was enclosed in a leak-proof aluminum foil blanket which was vented to the atmosphere through the wall of the autoclave to permit the escape of volatiles. Considerable outgassing occurred during the cure cycle. Figures 38 and 39 show the dense white gas (ammonia, phenol, water, and other reaction products) which issued from the exit end of the vent tube. Figure 39 was taken at the point in the cure cycle when pressure was applied. The sandwich composites were post cured under a blanket of argon to eliminate any oxygen degradation.

Heat aging, however, was done in air and it seems obvious that the post cured resin was still highly susceptible to oxygen attack at elevated temperatures. In fact, oxygen degradation was so severe at 600F that the specimens literally fell apart upon removal from

# TABLE XXXVIII

## PBI Sandwich Panels - 4.0 lb. Density Core
## Heat Aging Test Results

| Panel No. | Test Condition | Flatwise Tensile Str. | Compressive Str. | Flexural Shear Strength (Ribbon Direction) | Composite Bending Modulus (Ribbon Direction) | Flexural Shear Strength (Transverse Direction) | Composite Bending Modulus (Transverse Direction) |
|---|---|---|---|---|---|---|---|
| 65 | No Aging Tested at R. T. | 285 | 1259 | 406 | 4853 | 270 | 4160 |
| 90 | No Aging Tested at 400F | 382 | 268 | 181 | 2337 | 100 | 1408 |
| 92 | No Aging Tested at 500F | 295 | 251 | 189 | 1430 | 89 | 1224 |
| 93 | No Aging Tested at 600F | 265 | 187 | 203 | 2987 | 90 | 1072 |
| 86 | 100 Hrs at 500F Tested at 500F | 333 | 466 | 204 | 3570 | 93 | 2700 |
| 88 | 100 Hrs at 600F Tested at 600F | 46 | 110 | 82 | 2033 | 44 | 1153 |
| 84 | 300 Hrs at 400F Tested at 400F | 249 | 341 | 230 | 3433 | 93 | 2427 |
| 82 | 300 Hrs at 500F Tested at 500F | 439 | 334 | 212 | 3520 | 144 | 3213 |
| 81 | 300 Hrs at 600F Tested at 600F | Resin Burn Out | | | | | |
| 78 | 500 Hrs at 500F Tested at 500F | 135 | 218 | 179 | 3433 | 76 | 2533 |
| 66 | 500 Hrs at 600F Tested at 600F | Resin Burn Out | | | | | |
| 67 | 600 Hrs at 400F Tested at 400F | 187 | 290 | 223 | 3470 | 199 | 2743 |
| 68 | 600 Hrs at 500F Tested at 500F | 517 | 393 | 186 | 3403 | 73 | 2593 |
| 71 | 600 Hrs at 600F Tested at 600F | Resin Burn Out | | | | | |

## TABLE XXXIX

**PBI Sandwich Panels - 8.0 lb. Density Core**
**Heat Aging Test Results**

| Panel No. | Test Condition | Flat wise Tensile Str. | Compressive Str. | Flexural Shear Strength (Ribbon Direction) | Composite Bending Modulus (Ribbon Direction) | Flexural Shear Strength (Transverse Direction) | Composite Bending Modulus (Transverse Direction) |
|---|---|---|---|---|---|---|---|
| 89 | No Aging Tested at R. T. | 188 | 785 | 325 | 4683 | 254 | 4630 |
| 91 | No Aging Tested at 400F | 193 | 759 | 184 | 2117 | 200 | 2310 |
| 94 | No Aging Tested at 500F | 194 | 727 | 257 | 2874 | 114 | 1640 |
| 96 | No Aging Tested at 600F | 193 | 513 | 244 | 2637 | 207 | 2323 |
| 85 | 100 Hrs at 500F Tested at 500F | 128 | 512 | 321 | 4090 | 112 | 3380 |
| 87 | 100 Hrs at 600F Tested at 600F | 84 | 385 | 91.5 | 2973 | 106 | 2993 |
| 77 | 300 Hrs at 400F Tested at 400F | 358 | 405 | 345 | 4533 | 278 | 3810 |
| 80 | 300 Hrs at 500F Tested at 500F | 871 | 223 | 265 | 4287 | 252 | 3780 |
| 83 | 300 Hrs at 600F Tested at 600F | | | Resin | Burn | Out | |
| 72 | 500 Hrs at 500F Tested at 500F | 75 | 654 | 96 | 3317 | 119 | 3307 |
| 73 | 500 Hrs at 600F Tested at 600F | | | Resin | Burn | Out | |
| 74 | 600 Hrs at 400F Tested at 400F | 257 | 538 | 358 | 4173 | 289 | 3087 |
| 75 | 600 Hrs at 500F Tested at 500F | 539 | 419 | 146 | 4225 | 198 | 2880 |
| 76 | 600 Hrs at 600F Tested at 600F | | | Resin | Burn | Out | |

Figure 38 - Reaction Gases Flowing from Vent Tube During PBI Cure

Figure 39 - PBI Reaction Gases During Pressuring

the heat age oven. Figure 40 shows a series of 600F heat aged PBI sandwich specimens as they appeared at the end of their respective heat age periods. Figures 41, 42, 43, 44, and 45 show close up views of these heat aged series and give graphic evidence of the extent of the resin burn-out, adhesive degradation, and core deterioration.

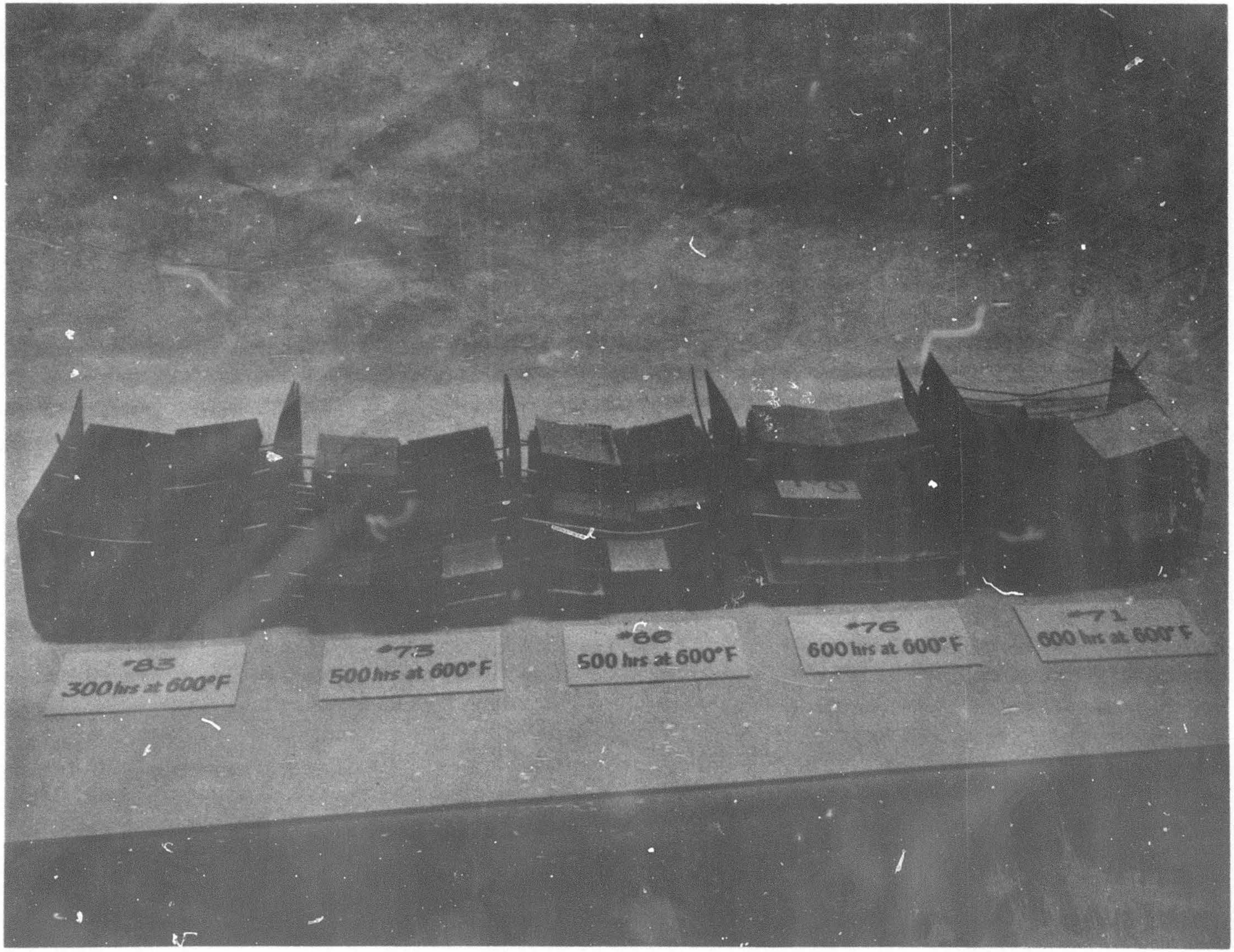



Figure 40 - PBI Sandwich Specimens after Heat Aging at 600 Degrees Fahrenheit

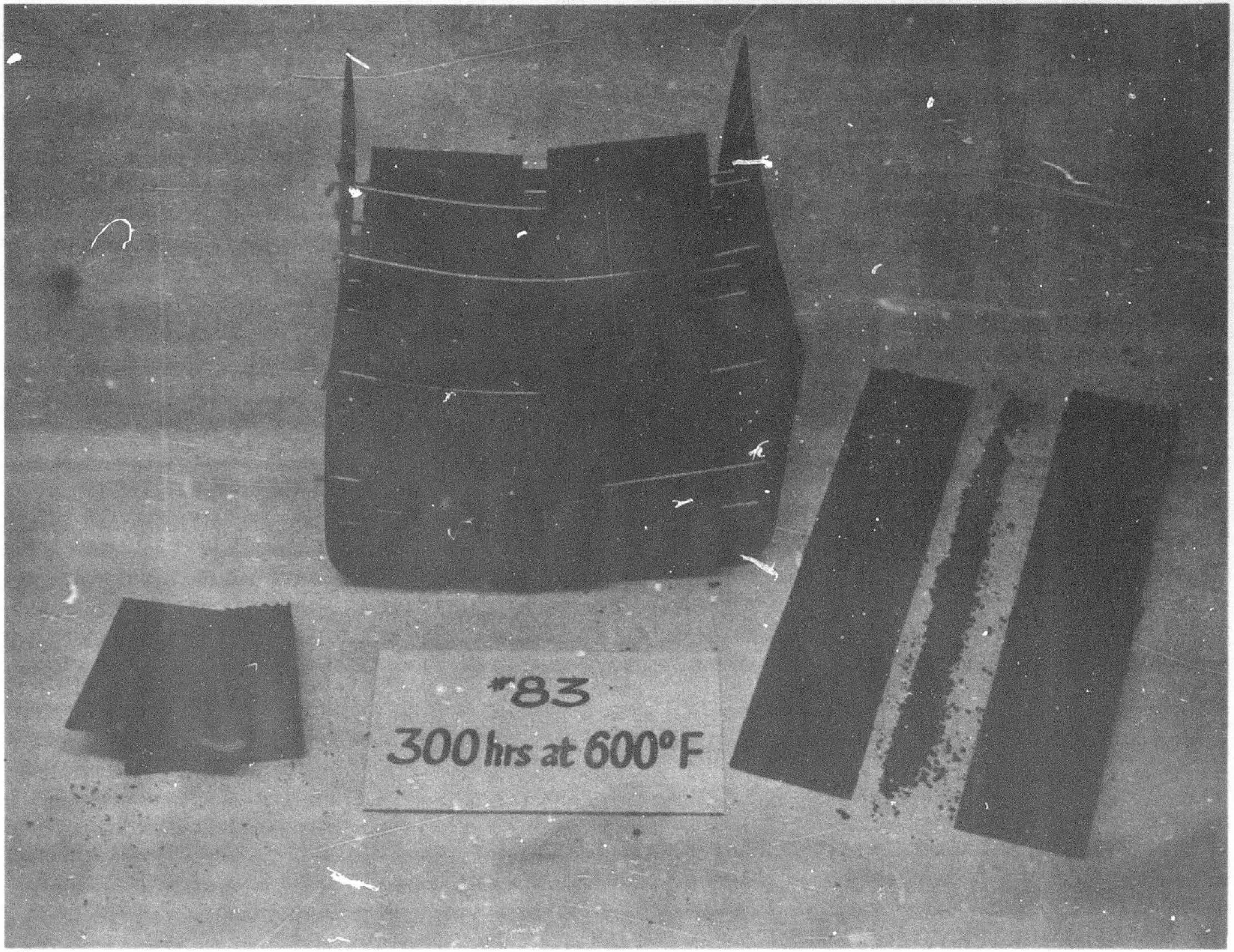



Figure 41 - PBI Specimens, 8.0 lb. Core - After 300 Hours at 600 Degrees Fahrenheit

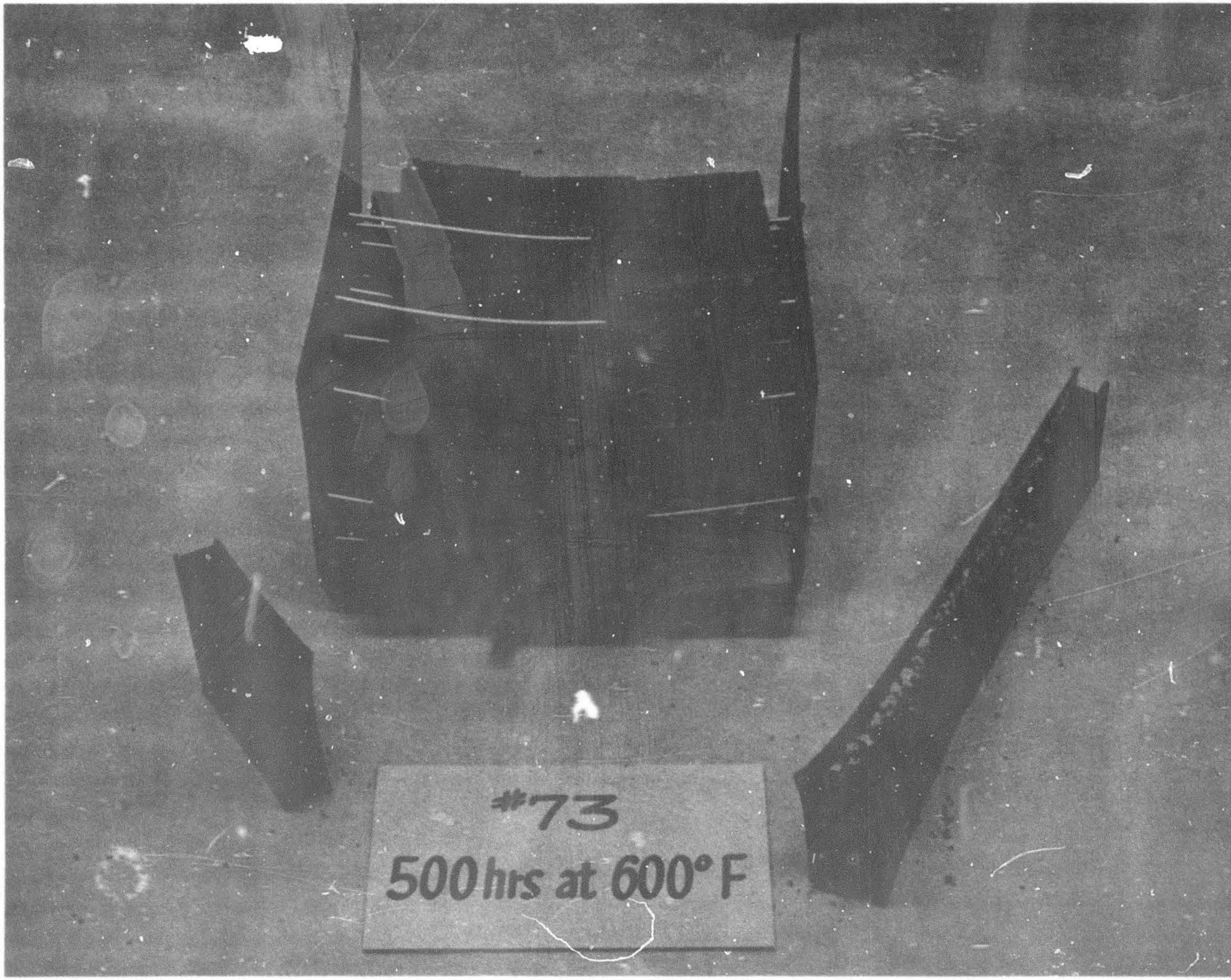



Figure 42 - PBI Specimens, 8.0 lb. Core - After 500 Hours at 600 Degrees Fahrenheit

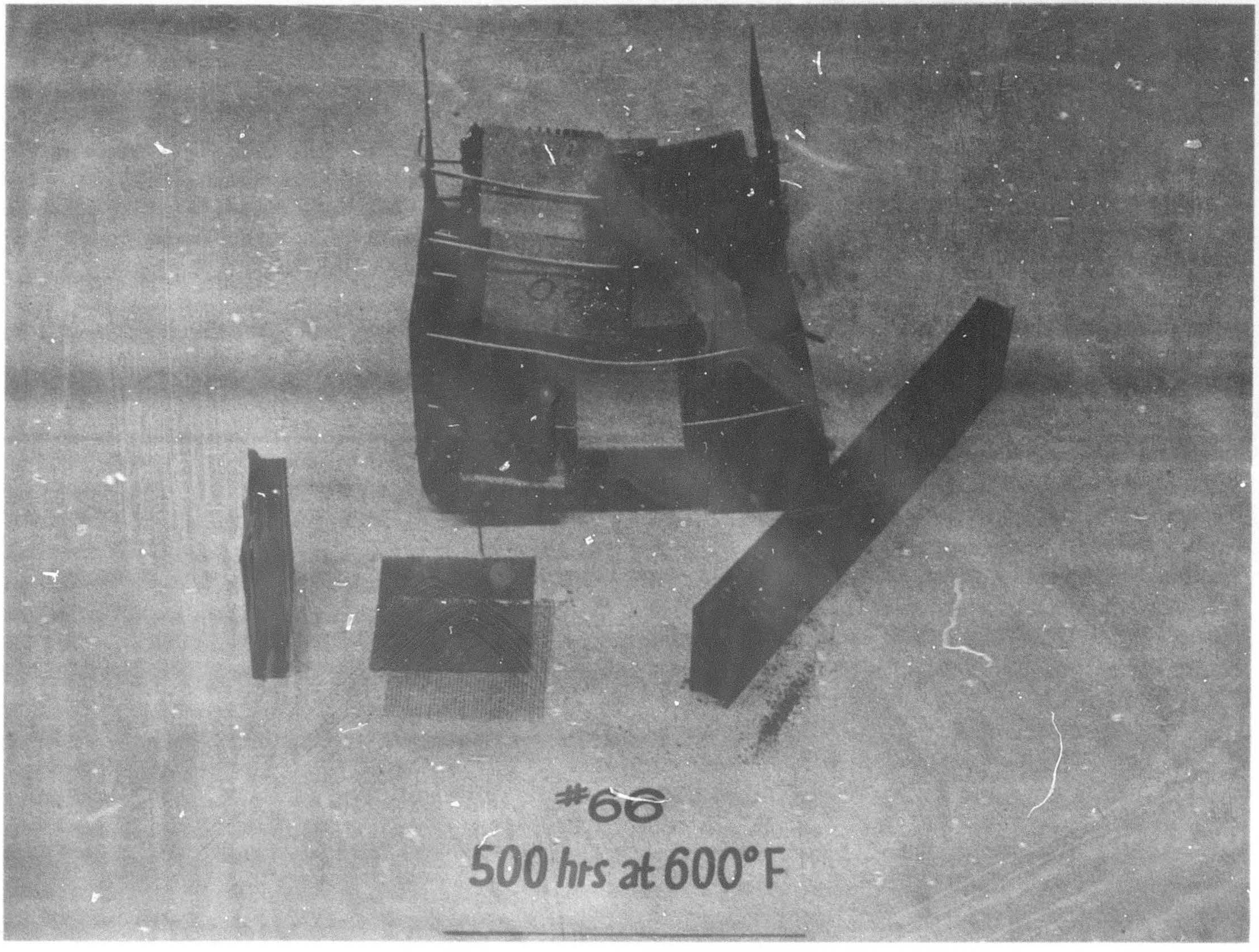



Figure 43 - PBI Specimens, 4.0 lb. Core - After 500 Hours at 600 Degrees Fahrenheit

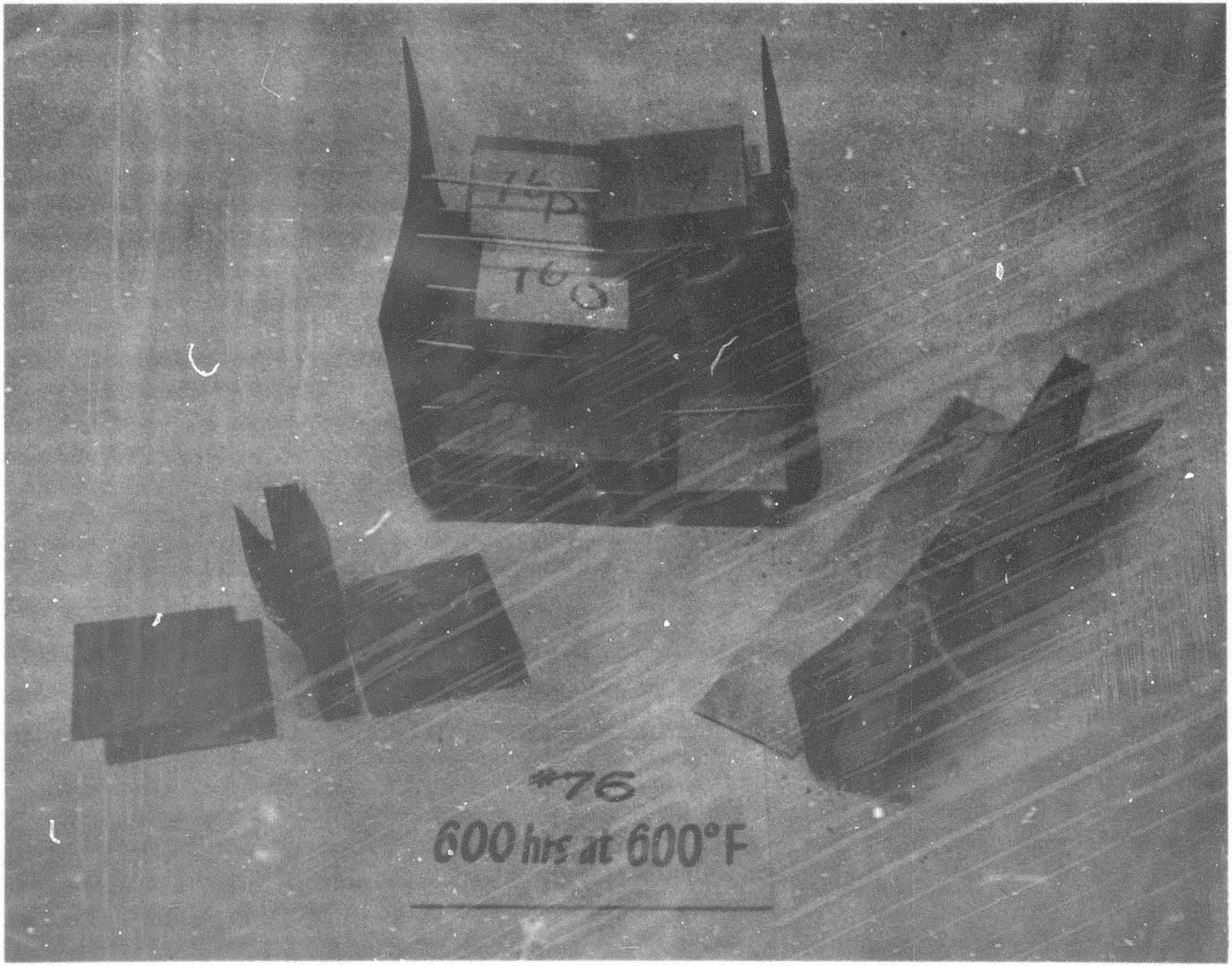



Figure 44 - PBI Specimens, 8.0 lb. Core - After 600 Hours at 600 Degrees Fahrenheit

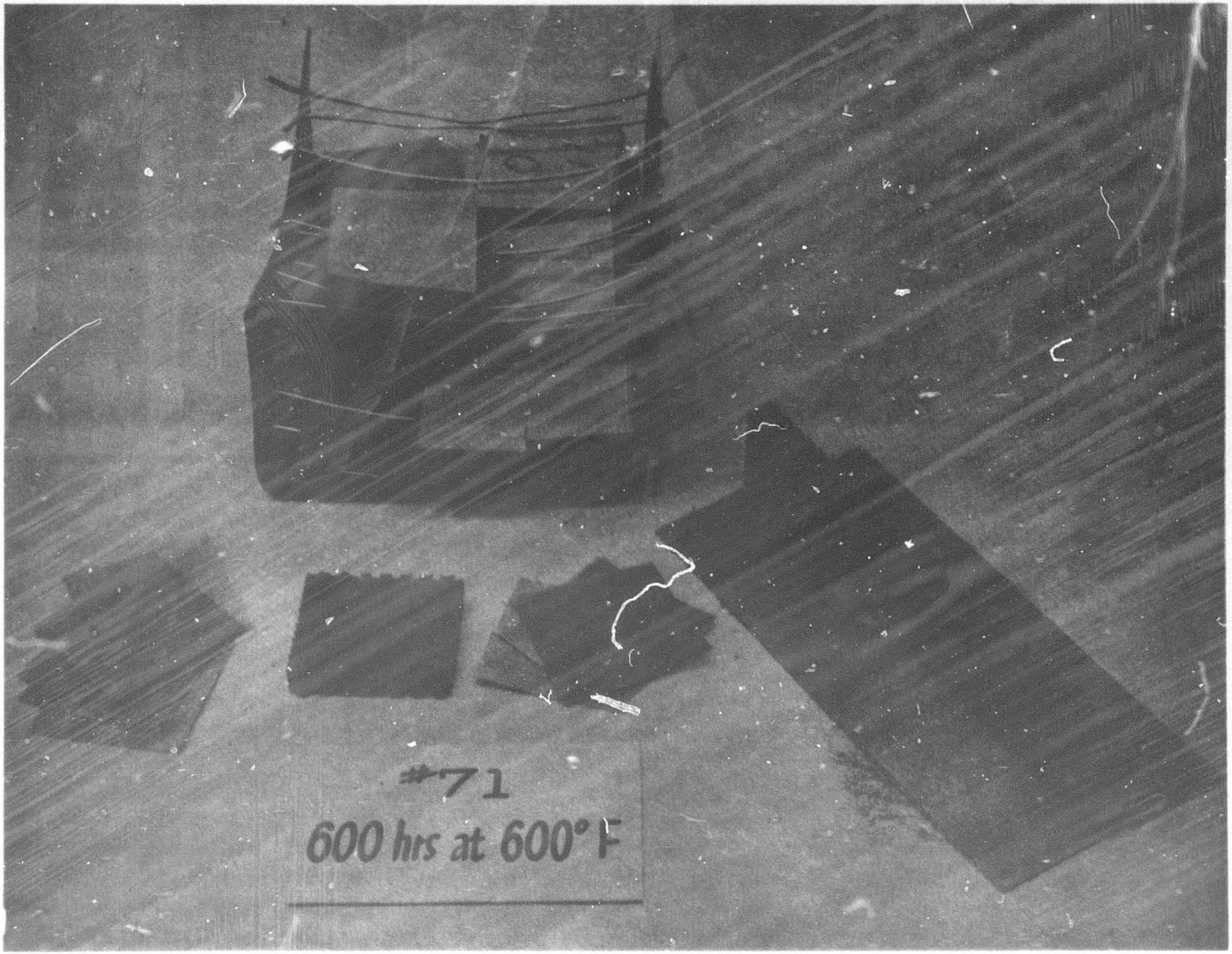



Figure 45 - PBI Specimens, 4.0 lb. Core - After 600 Hours at 600 Degrees Fahrenheit

# SECTION IX

# CORRELATION OF DATA - POLYIMIDE COMPOSITES

## A. GENERAL

The strength values for the polyimide sandwich composites remained high over the scheduled aging period of 600 hours for all test temperatures of 400F, 500F, and 600F. Heat aging tests were therefore continued. Values for shear strength and modulus in the ribbon direction were determined at 200-hour intervals beyond the 600-hour periods, according to the following schedule:

1. 4.0 lb. per cu. ft. density polyimide core

   a. 400F - Total of 1400 hours
   b. 500F - Total of 1400 hours

2. 8.0 lb. per cu. ft. density polyimide core

   a. 400F - Total of 1400 hours
   b. 500F - Total of 1200 hours

## B. GRAPHICAL PRESENTATION

The heat aging data collected on the polyimide honeycomb core sandwich composites were reduced to graphical form in the following manner.

1. An overall average room temperature value was obtained for the desired physical property of each sandwich configuration by using all appropriate room temperature values including the quality assurance and control values.

2. The quality assurance values obtained on each individual sample panel were considered the basic room temperature strengths for that panel. All test specimens were catalogued with reference to the sample panel from which they were cut.

3. The strength retention of each heat aged and tested specimen was determined by relating back to the basic room temperature values of the panel from which the specimen was cut.

4. The Heat Age graphs were drawn to show "Percent Retention of Room Temperature Strength" versus "Hours of Heat Aging".

5. The Overall Average Room Temperature Strength was printed on each graph.

The Heat Age Graphs on polyimide sandwich composites presented in this report are:

Figure 46 - Flatwise Tensile Strength - 4.0 lb. Density Core
Figure 47 - Compressive Strength - 4.0 lb. Density Core
Figure 48 - Flexural Shear Strength - Ribbon Direction - 4.0 lb Core
Figure 49 - Composite Bending Modulus - Ribbon Direction - 4.0 lb. Core
Figure 50 - Flexural Shear Strength - Transverse Direction - 4.0 lb. Core
Figure 51 - Composite Bending Modulus - Transverse Direction - 4.0 lb. Core
Figure 52 - Flatwise Tensile Strength - 8.0 lb. Density Core
Figure 53 - Compressive Strength - 8.0 lb. Density Core
Figure 54 - Flexural Shear Strength - Ribbon Direction - 8.0 lb. Core
Figure 55 - Composite Bending Modulus - Ribbon Direction - 8.0 lb. Core
Figure 56 - Flexural Shear Strength - Transverse Direction - 8.0 lb. Core
Figure 57 - Composite Bending Modulus - Transverse Direction - 8.0 lb. Core

The figures present a graphical picture of the changes in the physical strengths of the polyimide composites during heat aging at 400F, 500F, and 600F.

## C. DISCUSSION OF HEAT AGE DATA

1. The heat resistance of both the 4.0 lb. density core composite and the 8.0 lb. density core composite with respect to all the physical properties tested was excellent. The polyimide resin showed outstanding high-temperature strength properties at 400F, 500F, and 600F.

2. It should be noted that even after 1400 hours aging at 400F and at 500F, the shear strength and modulus of the 4.0 lb. density core construction remained well above 50% of the room temperature values. (Figure 48). After 1000 hours there appeared to be a slight decline in strength. However, the values at the 1400-hour mark were nearly the same as the initial values before heat aging started. It was, therefore, difficult to tell whether the apparent downward trend was actual or only circumstantial. It was obvious that the useful life of the composite had not by any means been reached. It was felt that if design values for the 4.0 lb. density core polyimide sandwich composite were to

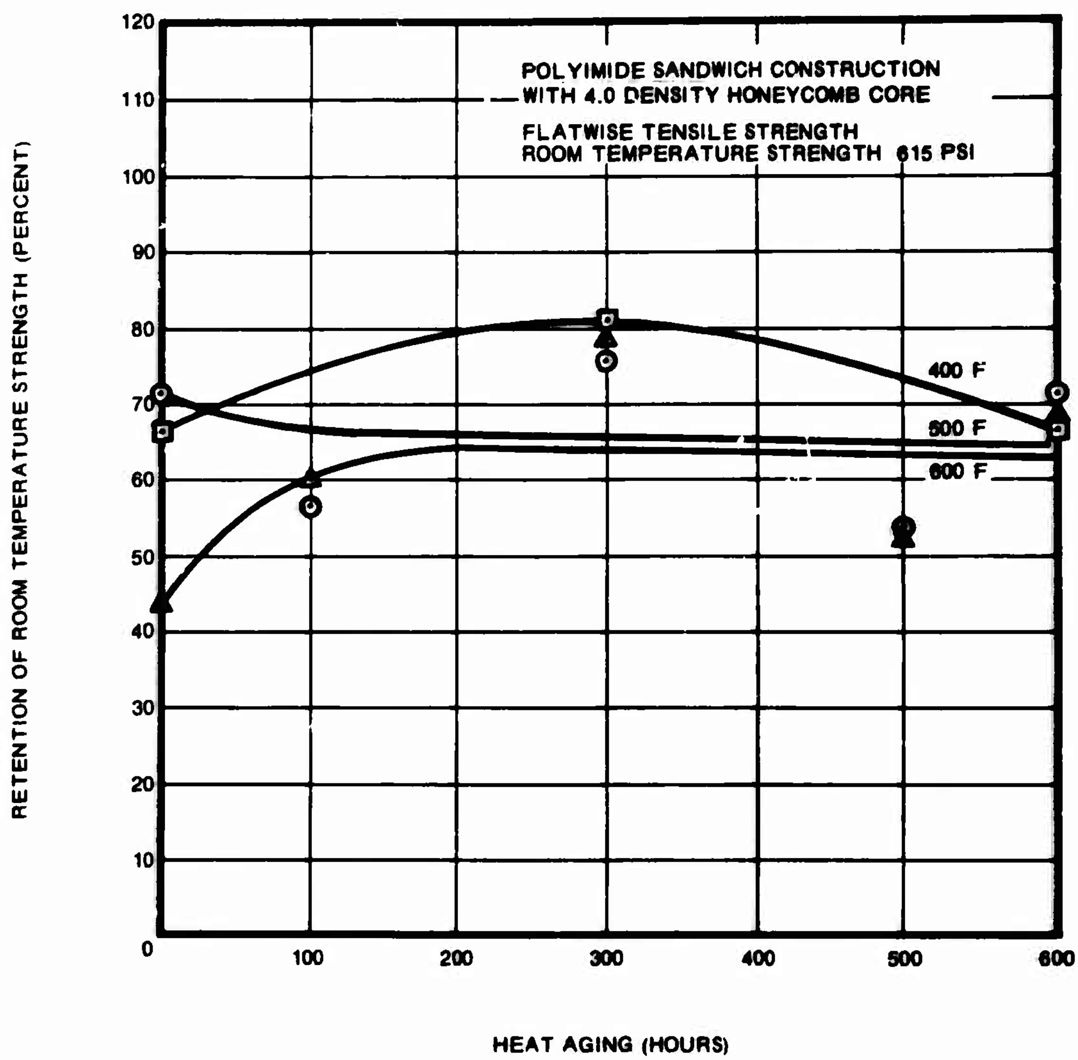



Figure 46 - Polyimide Composite, 4.0-lb Core, Flatwise Tensile Strength, Hours Aging versus Percent Retention of Room Temperature Strength

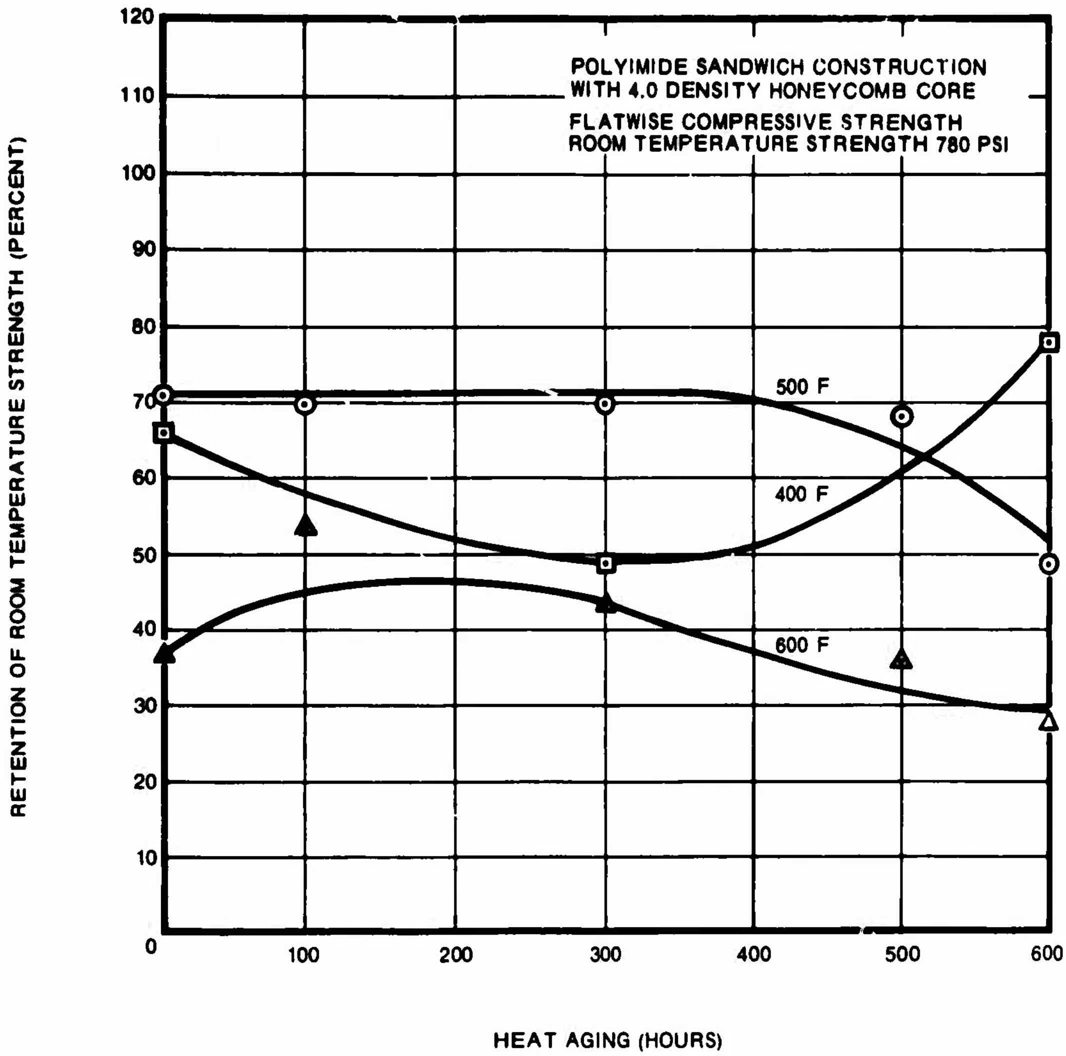



Figure 47 - Polyimide Composite, 4.0-lb Core, Compressive Strength, Hours Aging versus Percent Retention of Room Temperature Strength

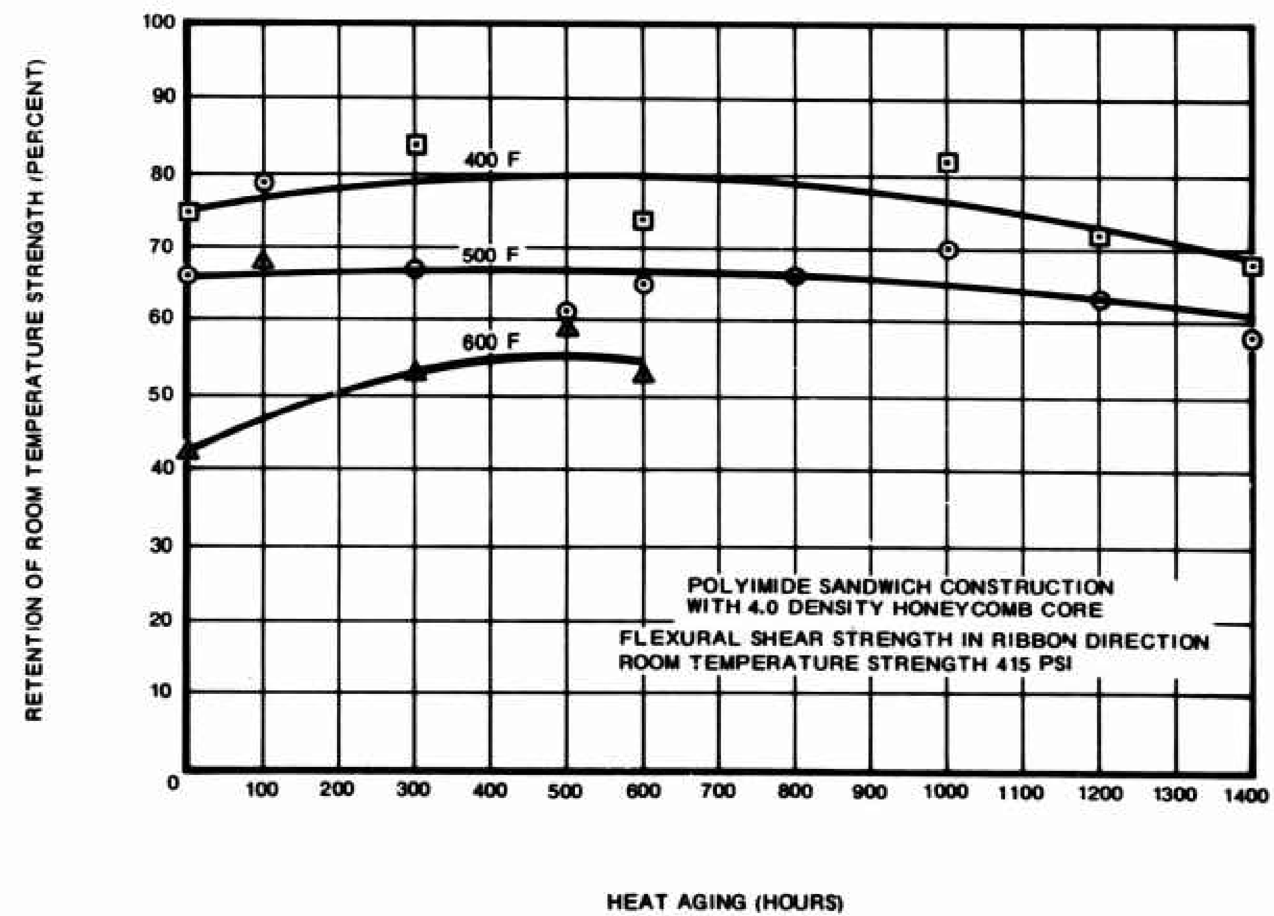



**Figure 48 - Polyimide Composite, 4.0-lb Core, Ribbon Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength**

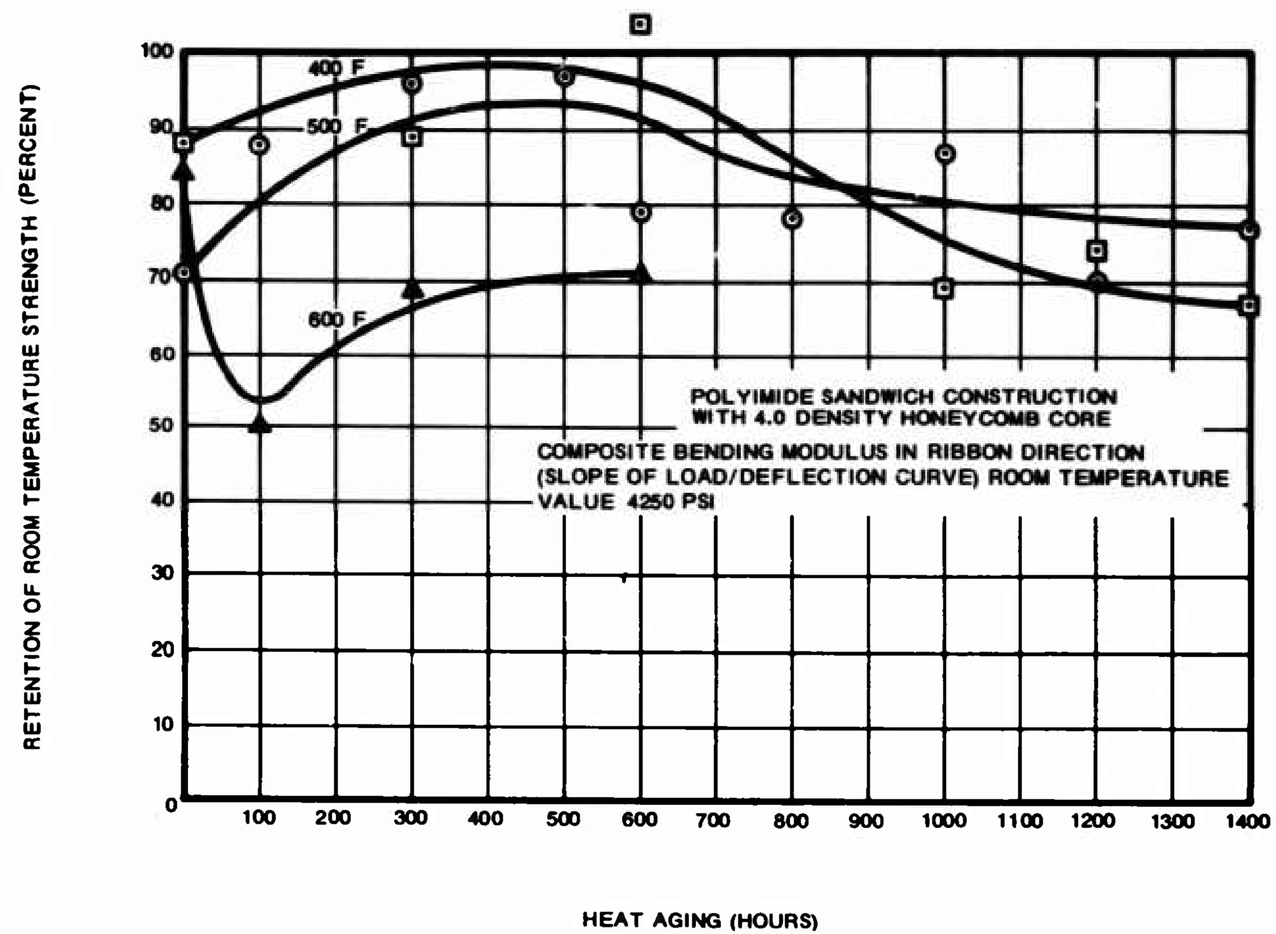



Figure 49 - Polyimide Composite, 4.0-lb Core, Ribbon Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength

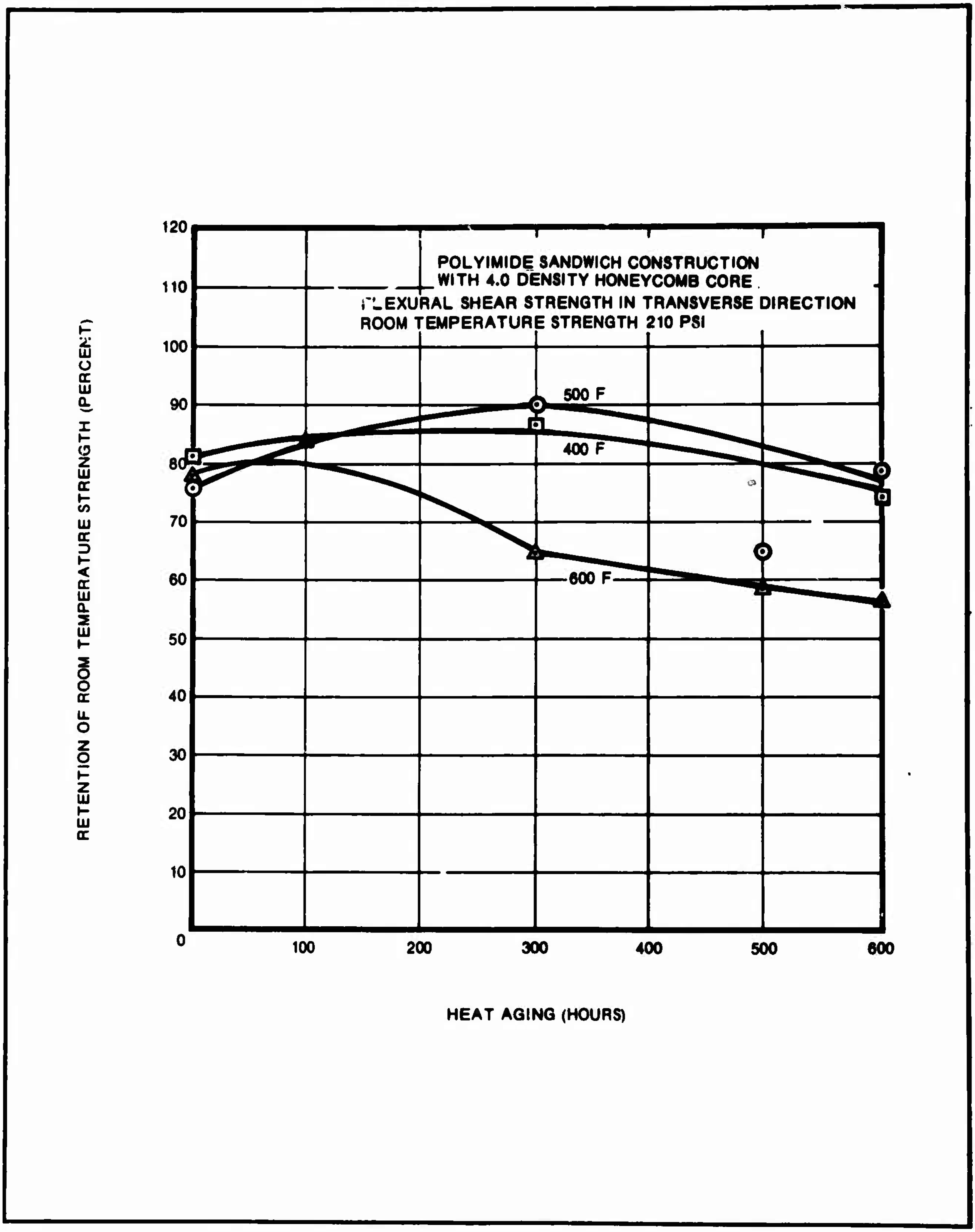



Figure 50 - Polyimide Composite, 4.0-lb Core, Transverse Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

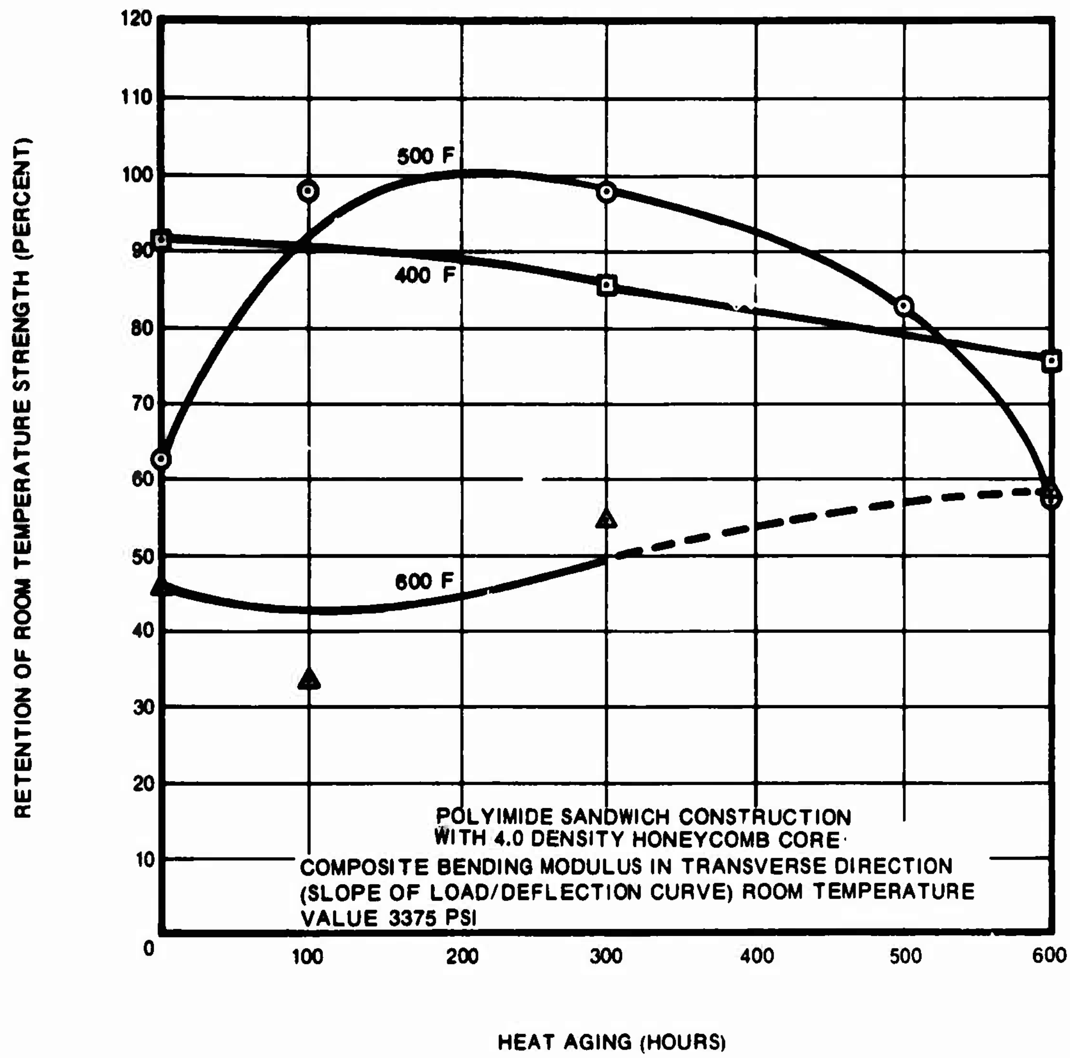



Figure 51 - Polyimide Composite, 4.0-lb Core, Transverse Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength

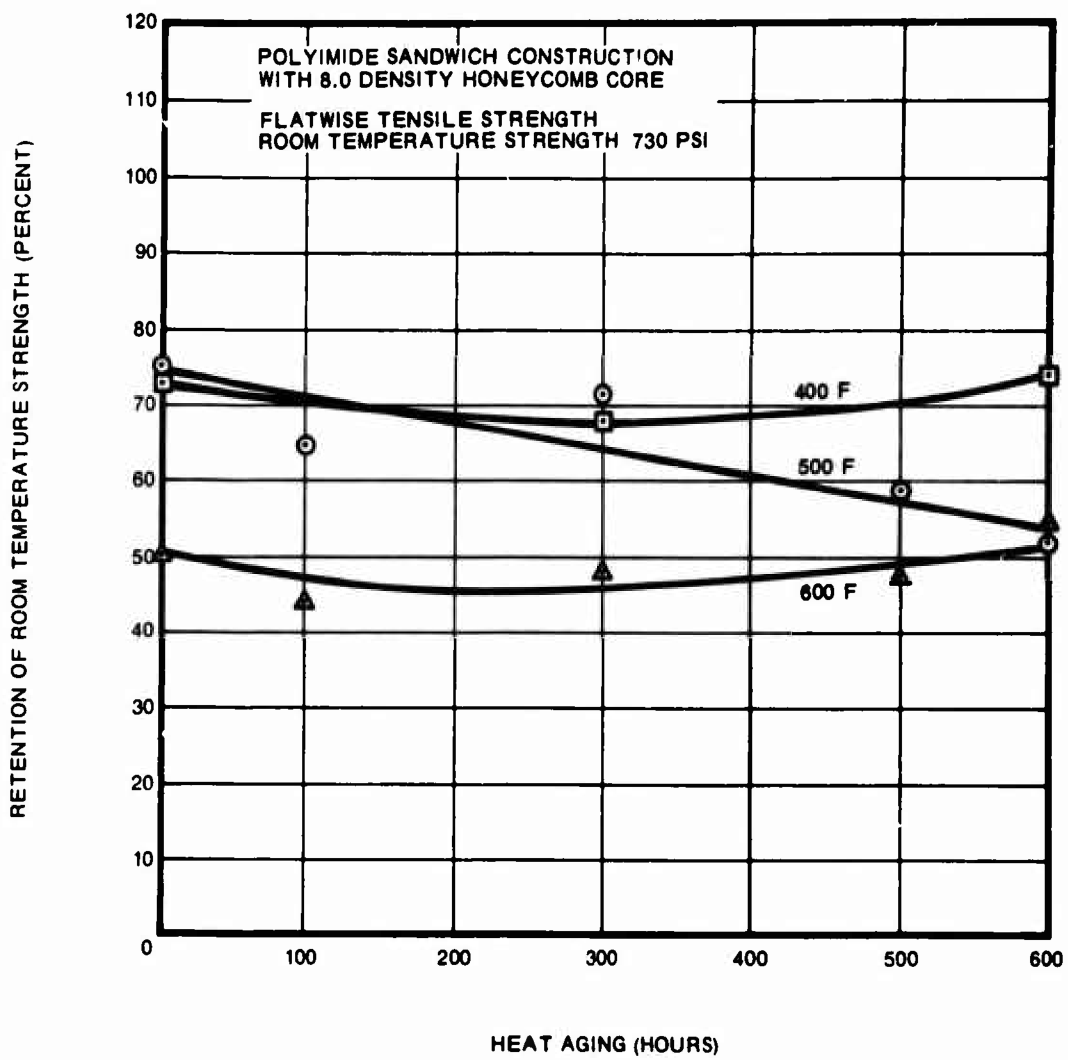



Figure 52 - Polyimide Composite, 8.0-lb Core, Flatwise Tensile Strength, Hours Aging versus Percent Rete... Room Temperature Strength

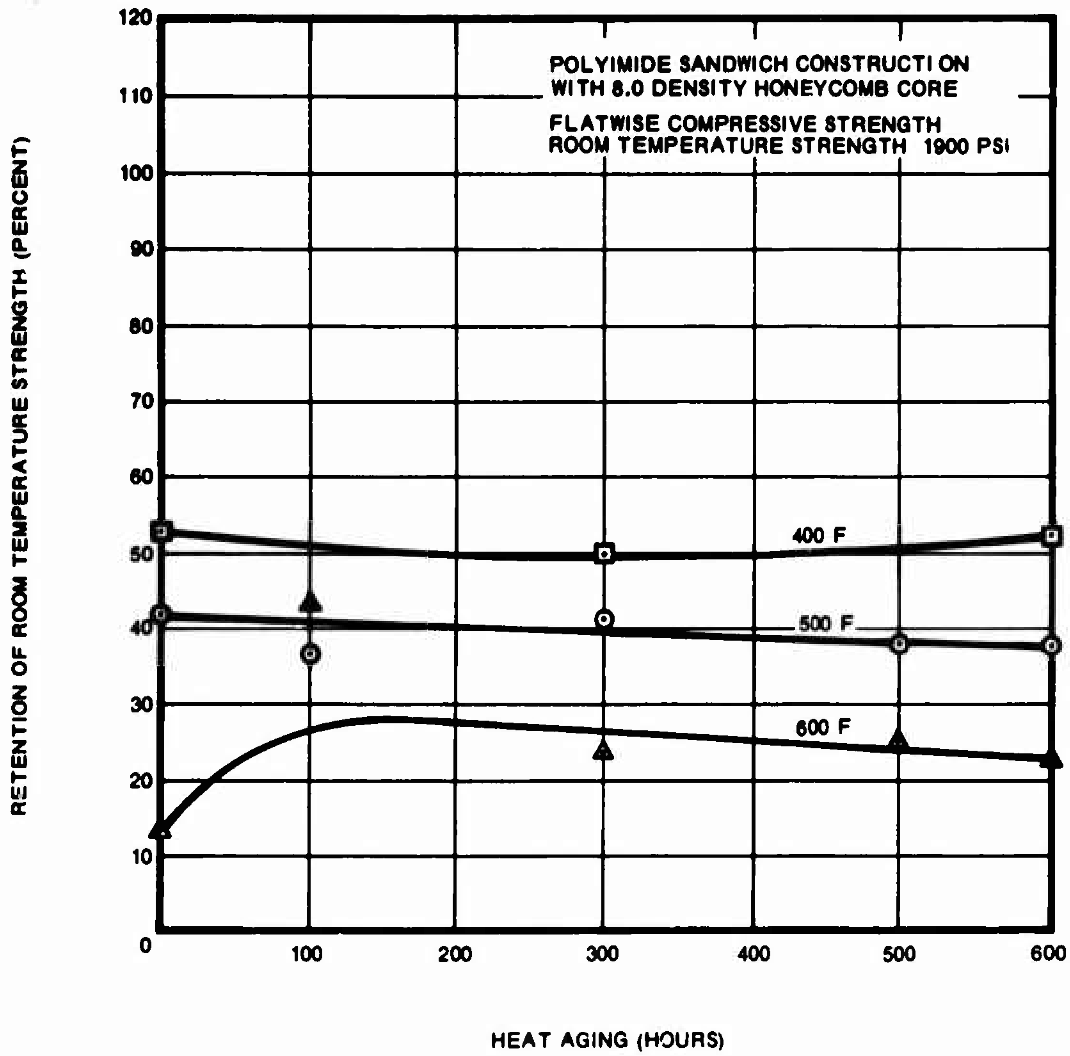



Figure 53 - Polyimide Composite, 8.0-lb Core, Compressive Strength, Hours Aging versus Percent Retention of Room Temperature Strength

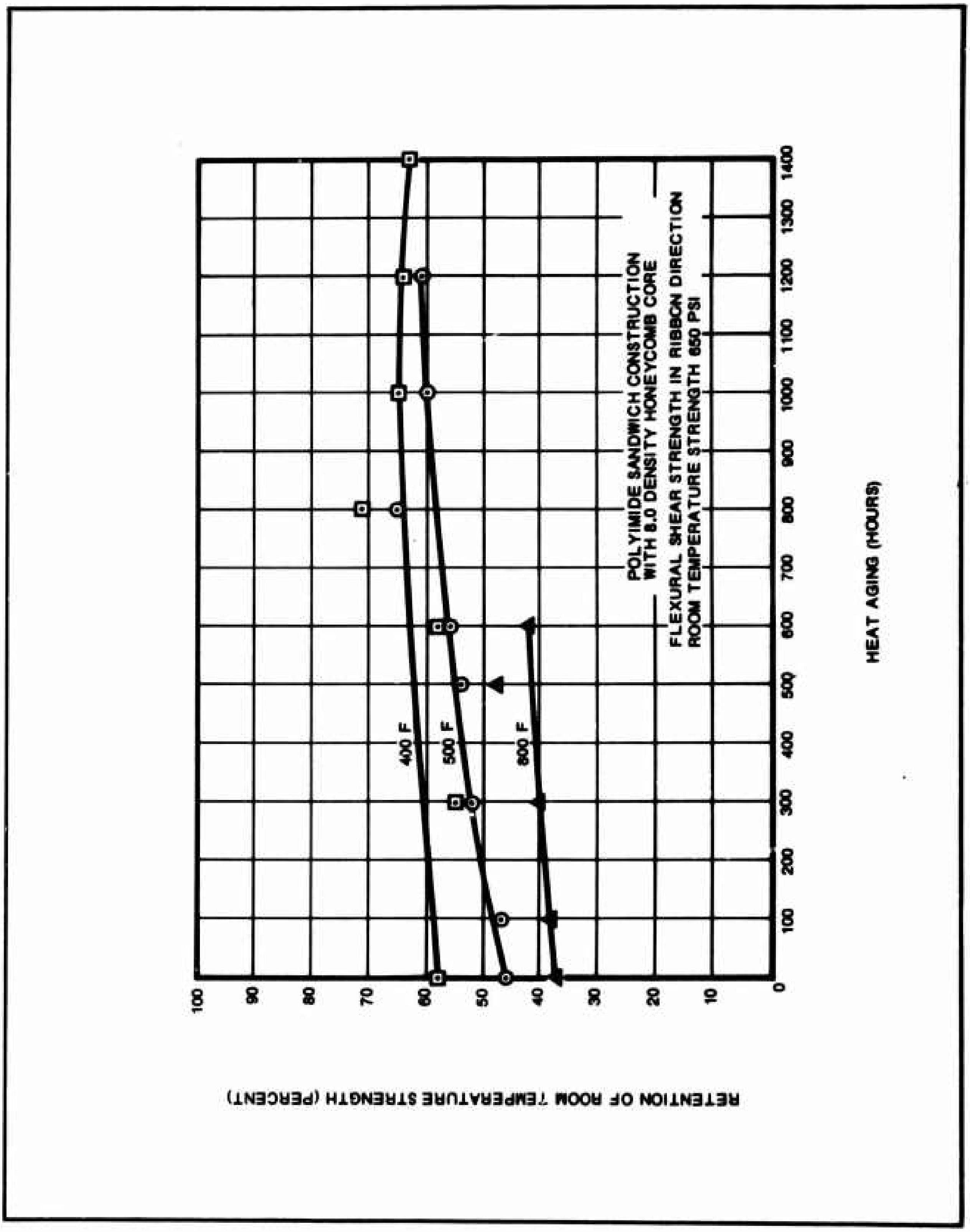



Figure 54 - Polyimide Composite, 8.0-lb Core, Ribbon Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

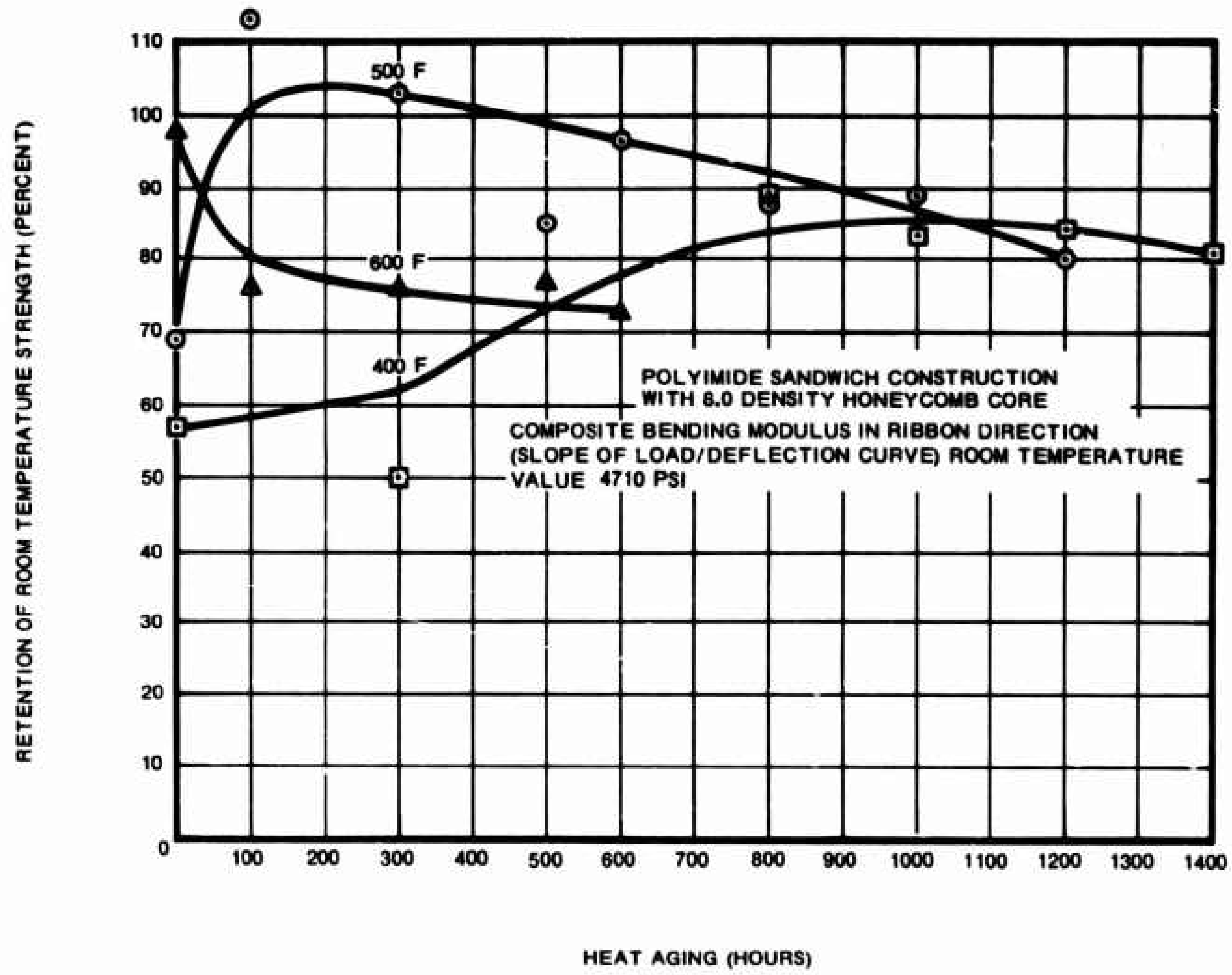



Figure 55 - Polyimide Composite, 8.0-lb Core, Ribbon Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength

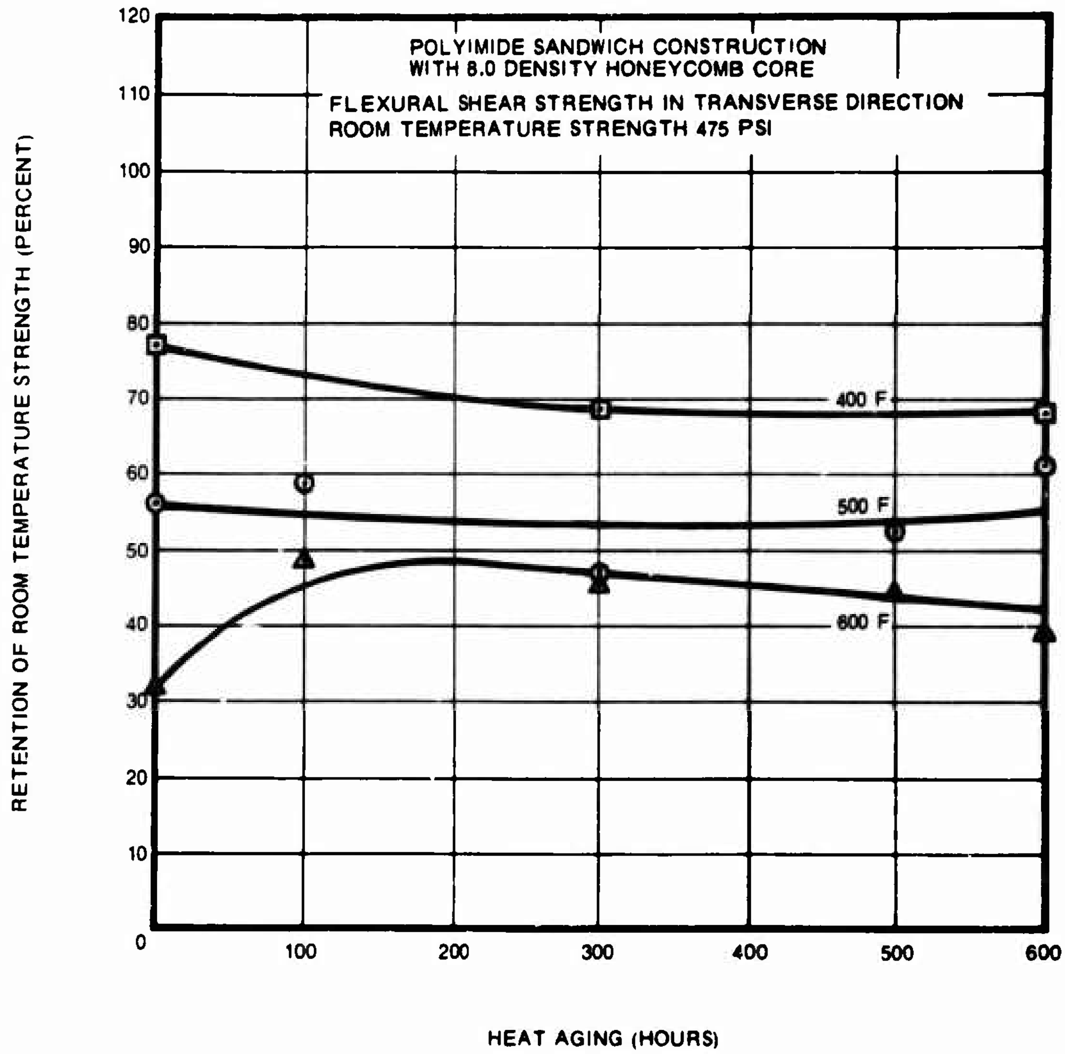



Figure 56 - Polyimide Composite, 8.0-lb Core, Transverse Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

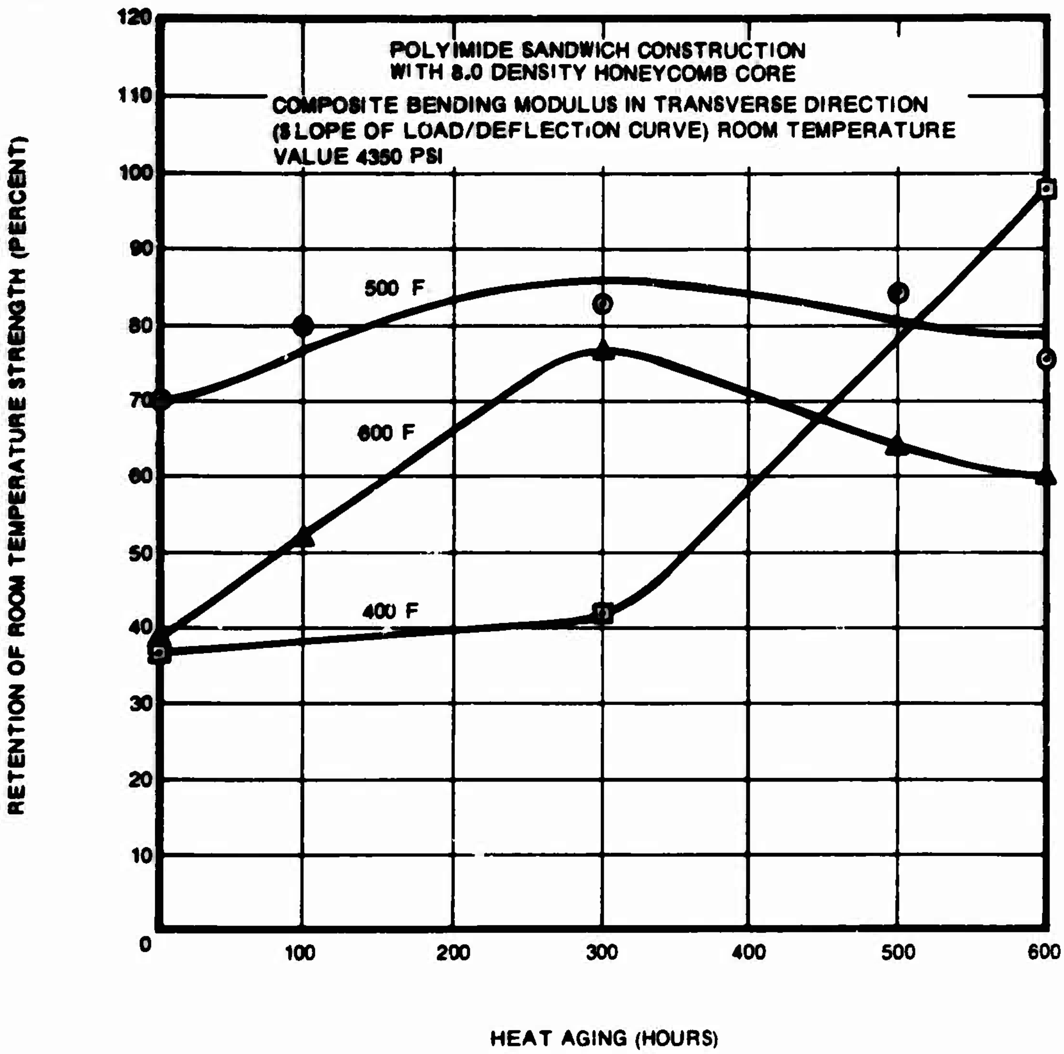



Figure 57 - Polyimide Composite, 8.0-lb Core, Transverse Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength

be determined for extended use at elevated temperatures, the heat aging tests would have to be carried out beyond the 5000-hour mark and probably to 10,000 hours. More valid design values would also be obtained if the test procedures were more rigorous. Testing should go beyond simple static heat aging and include heat aging under stress, fatigue testing, and cyclic testing.

3. It is quite apparent that the comments made in the preceding paragraph concerning the useful life of the 4.0 lb. core density polyimide composites are equally true for the 8.0 lb. core density polyimide composites. The final values at the end of 1400 hours aging at 400F and 1200 hours aging at 500F on the 8.0 lb. core density composites were actually higher than the initial values before heat aging began. (Figure 54). The useful life of the composites most certainly lay well beyond these aging periods.

4. The 8.0 lb. density core composite was stronger in every respect than the 4.0 lb. density core composite based on room temperature strength values.

5. When tested at elevated temperatures (no aging), the 8.0 lb. core composite was stronger based on actual strength values, but the 4.0 lb. core composite appeared to retain a higher per cent of its room temperature strength. This was probably due to the fact that in fiberglass reinforced composites the lower the resin binder (within limits) the more predominant becomes the high temperature strength of the glass reinforcement.

6. The 8.0 lb. core composite appeared to be slightly more heat resistant than the 4.0 lb. composite. The strength values for the 8.0 composite were more uniform and more stable. However, the heat resistant properties of both composites were so outstanding over the aging periods involved that it is difficult to determine which core has superior heat resistant properties. Longer aging periods would be required to make a firm distinction between the two cores.

## CORRELATION OF DATA - PBI COMPOSITES

### A. GENERAL

Test values for the PBI composites were variable and unpredictable. The susceptibility of the PBI resin to oxygen degradation during cure, post cure, and heat aging coupled with the often extreme non-uniformity of the prepreg, adhesive film, and honeycomb core gave the fabrication and testing program on PBI honeycomb composites a high degree of variation.

All specimens aged at 600F experienced complete failure before the end of the 300 hour test period. Specimens aged at 500F exhibited definite signs of degradation. Strength values in most cases had dropped below 50% of room temperature values after 600 hours aging.

All specimen configurations aged at 400F retained over 50% of their room temperature strength at the 600 hour test period. Additional testing would be required to definitely establish the heat resistance capabilities of the PBI composite at 400F. Time did not permit extension of the tests beyond the 600-hour period, however.

### B. GRAPHICAL PRESENTATION

The heat aging data collected on the PBI (2803) honeycomb core sandwich composites were reduced to graphical form in the manner described in Section IX-B (on polyimide composites).

The Heat Age graphs on PBI sandwich composites presented in this report are:

Figure 58 - Flatwise Tensile Strength - 4.0 lb. Density Core
Figure 59 - Compressive Strength - 4.0 lb. Density Core
Figure 60 - Flexural Shear Strength - Ribbon Direction - 4.0 lb. Core
Figure 61 - Composite Bending Modulus - Ribbon Direction 4.0 lb. Core
Figure 62 - Flexural Shear Strength - Transverse Direction - 4.0 lb. Core
Figure 63 - Composite Bending Modulus - Transverse Direction - 4.0 lb. Core
Figure 64 - Flatwise Tensile Strength - 8.0 lb. Density Core
Figure 65 - Compressive Strength - 8.0 lb. Core
Figure 66 - Flexural Shear Strength - Ribbon Direction - 8.0 lb. Core

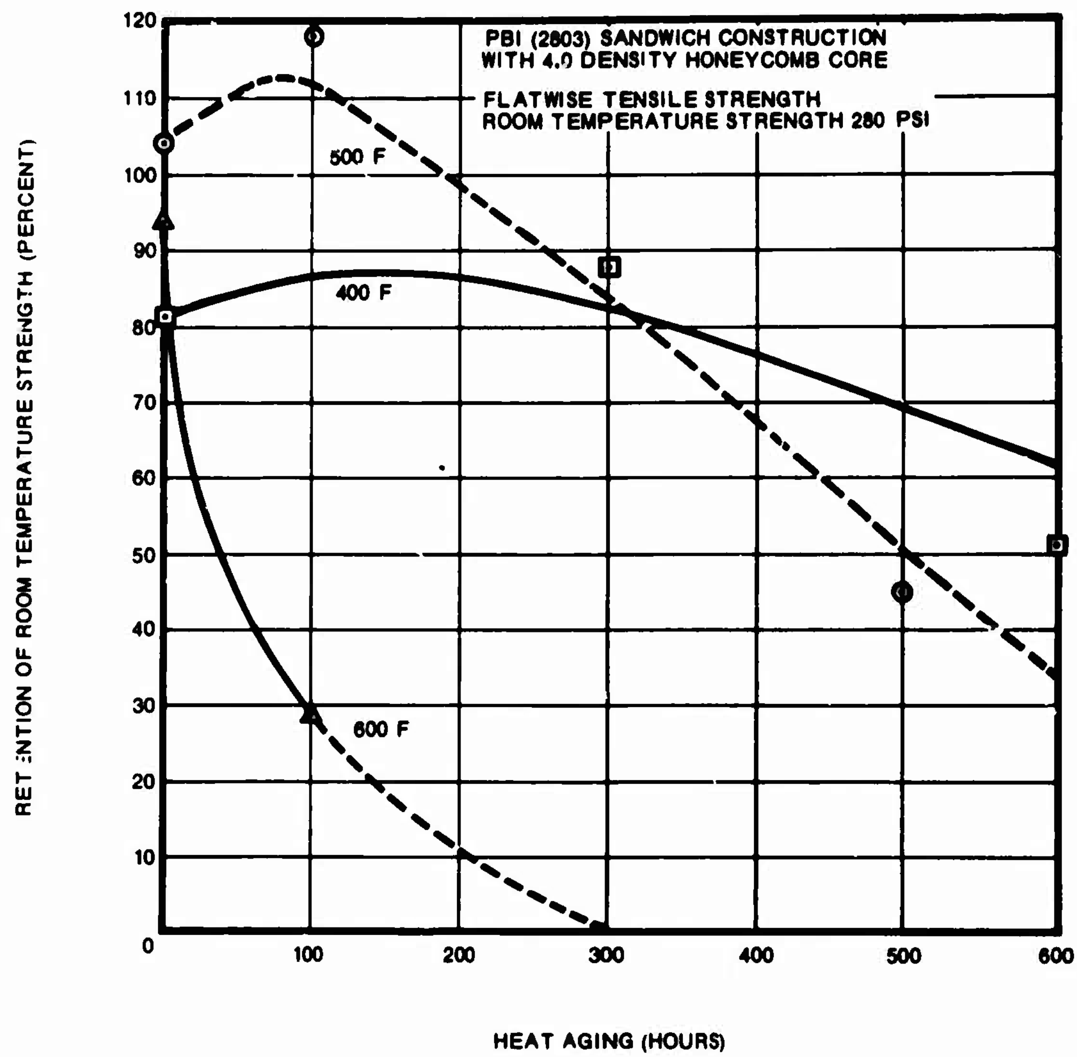



Figure 58 - PBI Composite, 4.0-lb Core, Flatwise Tensile Strength, Hours Aging versus Percent Retention of Room Temperature Strength

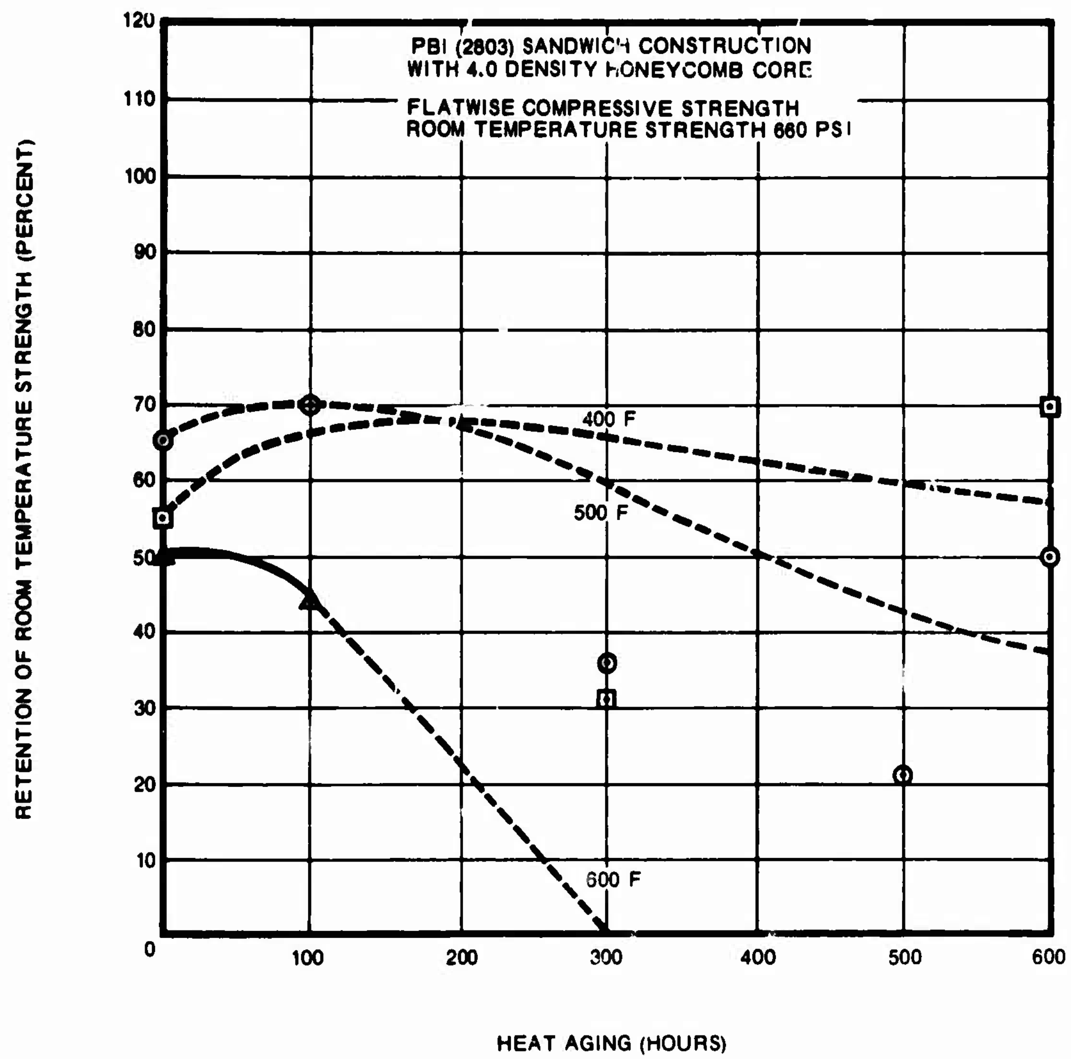



Figure 59 - PBI Composite, 4.0-lb Core, Compressive Strength, Hours Aging versus Percent Retention of Room Temperature Strength

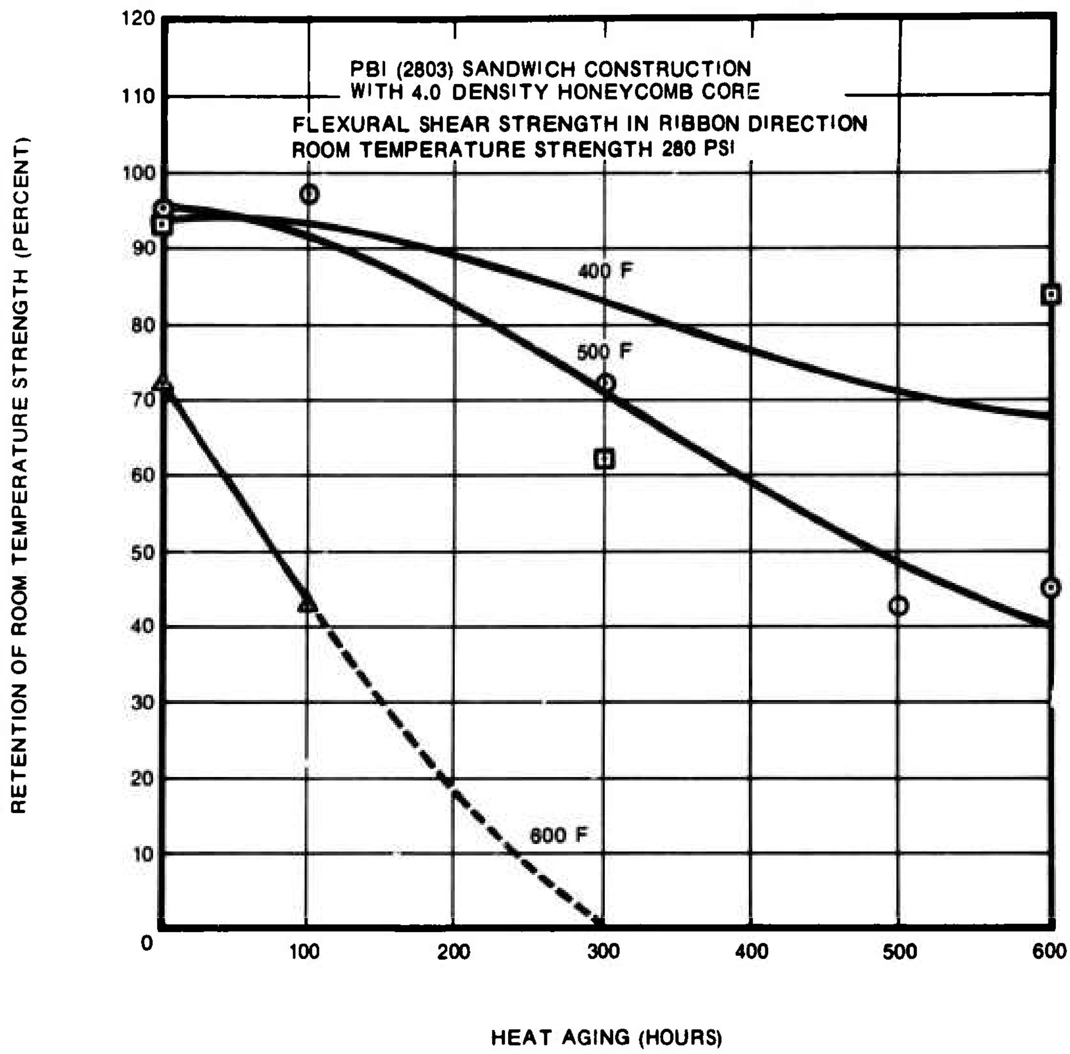



Figure 60 - PBI Composite, 4.0-lb Core, Ribbon Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

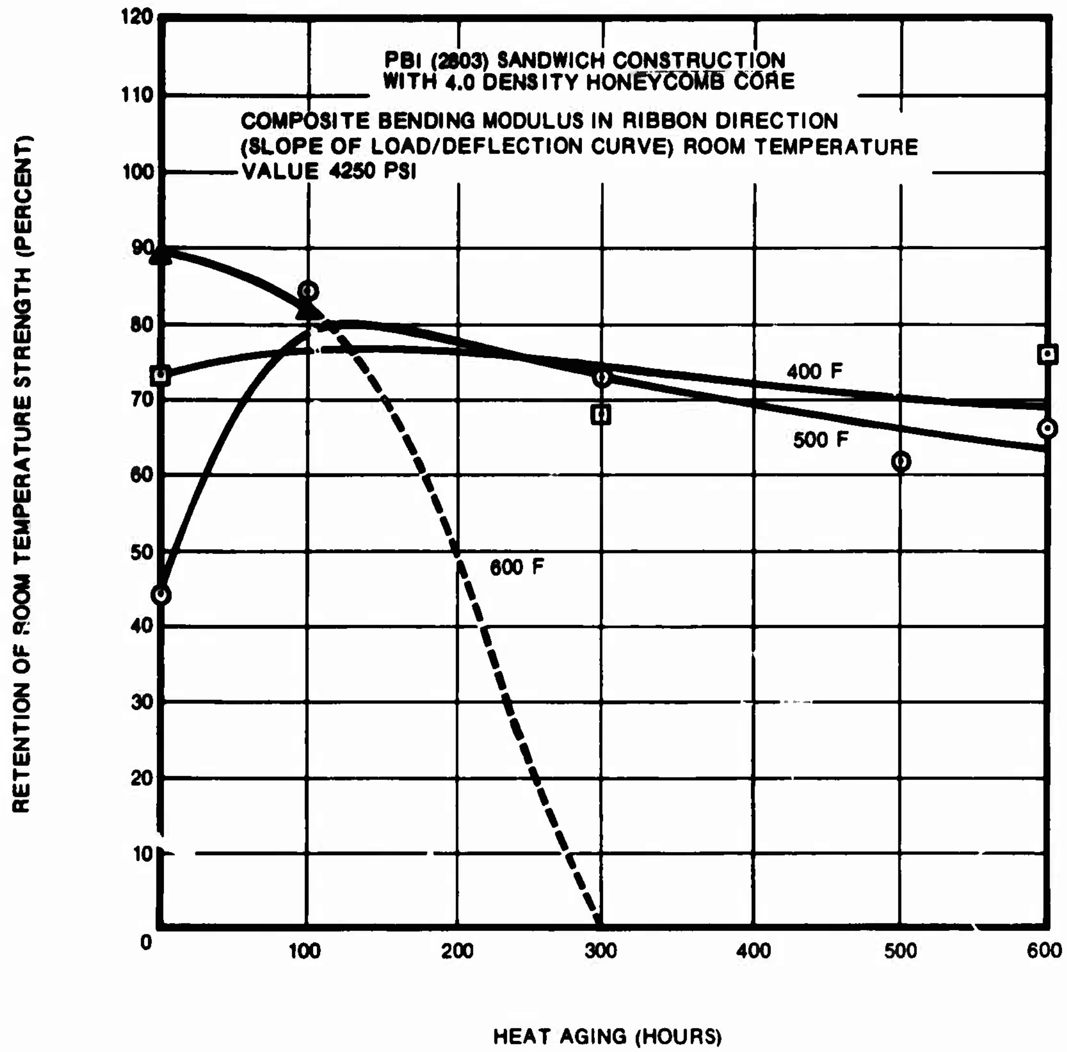



**Figure 61 - PBI Composite, 4.0-lb Core, Ribbon Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength**

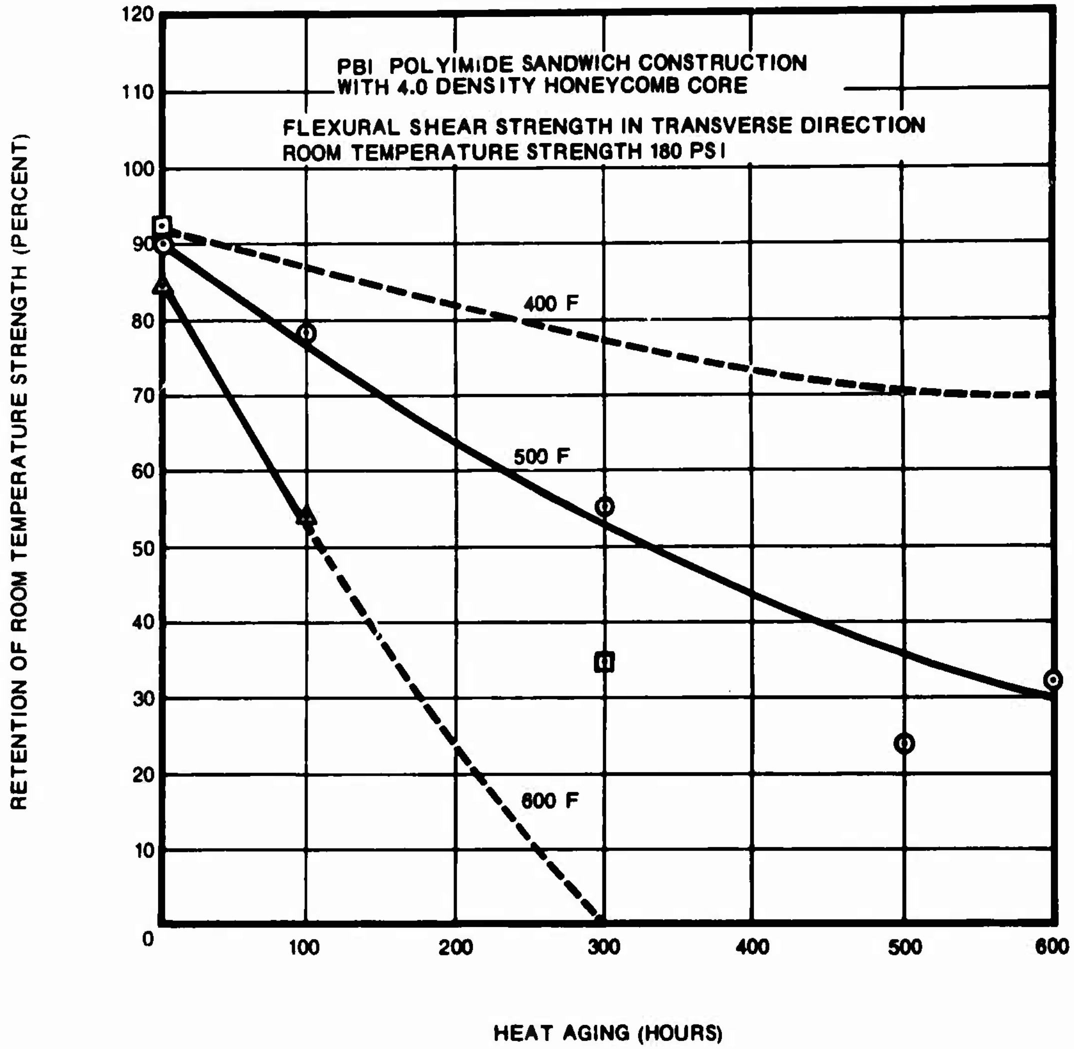



Figure 62 - PBI Composite, 4.0-lb Core, Transverse Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

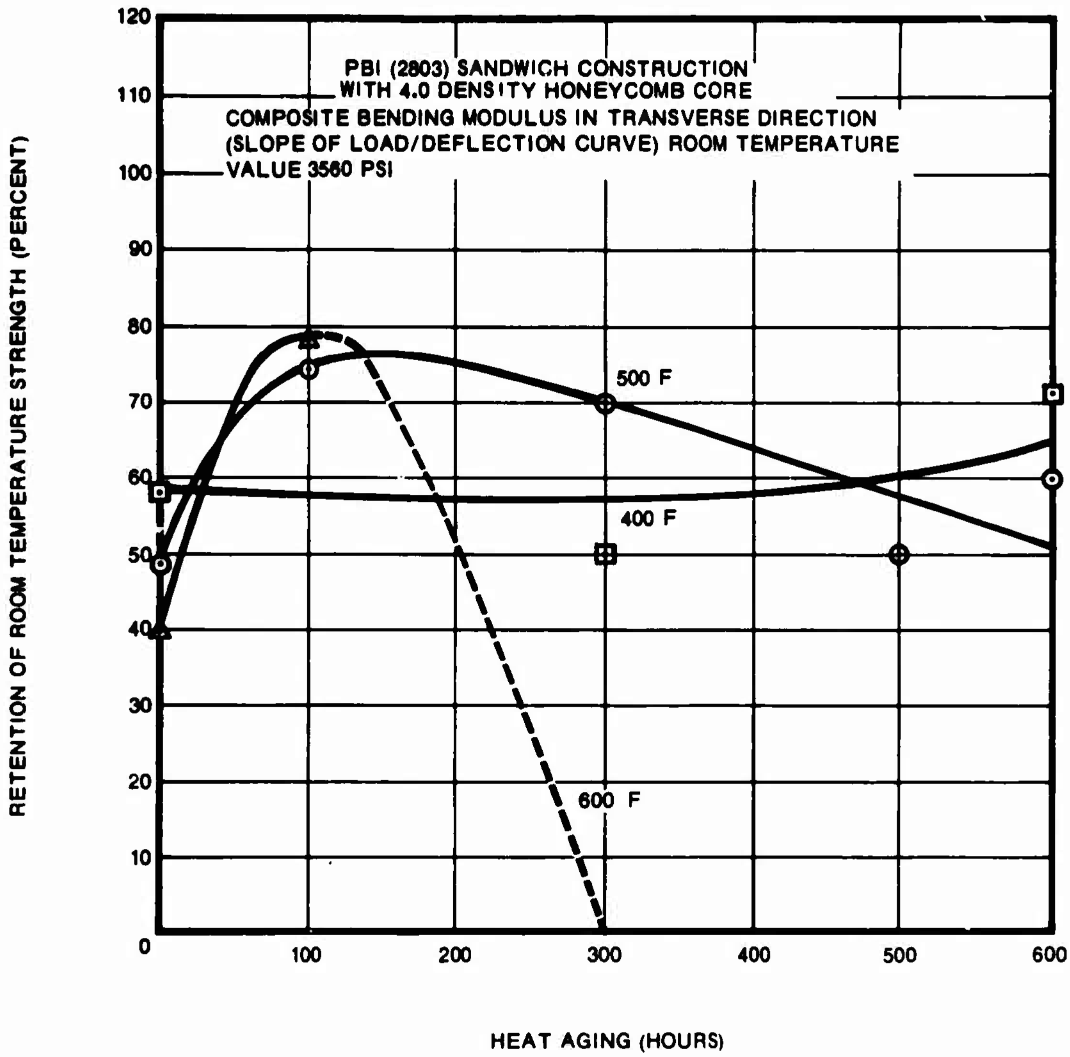



Figure 63 - PBI Composite, 4.0-lb Core, Transverse Composite Bending Modulus, Hours Aging versus Percent Retention oi Room Temperature Strength

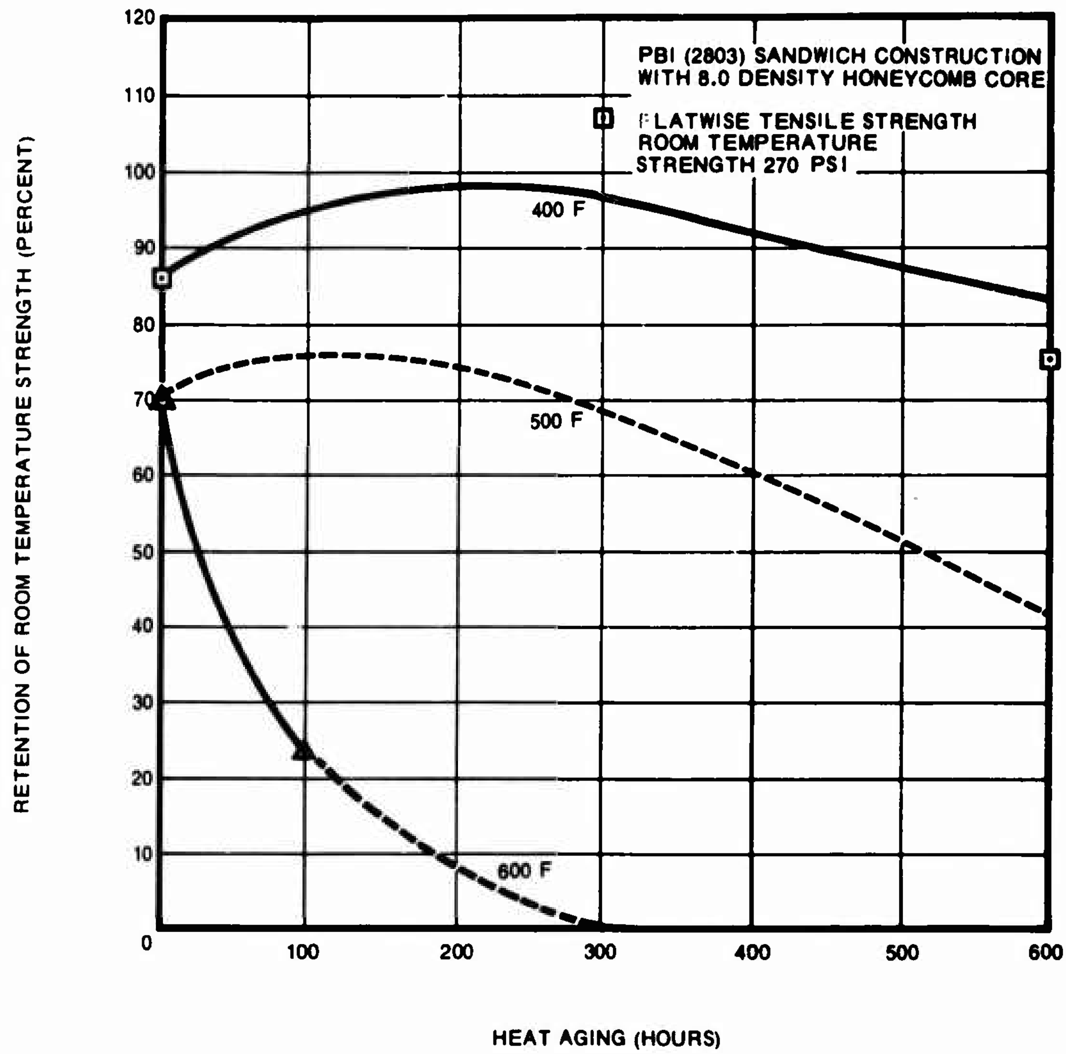



Figure 64 - PBI Composite, 8.0-lb Core, Flatwise Tensile Strength, Hours Aging versus Percent Retention of Room Temperature Strength

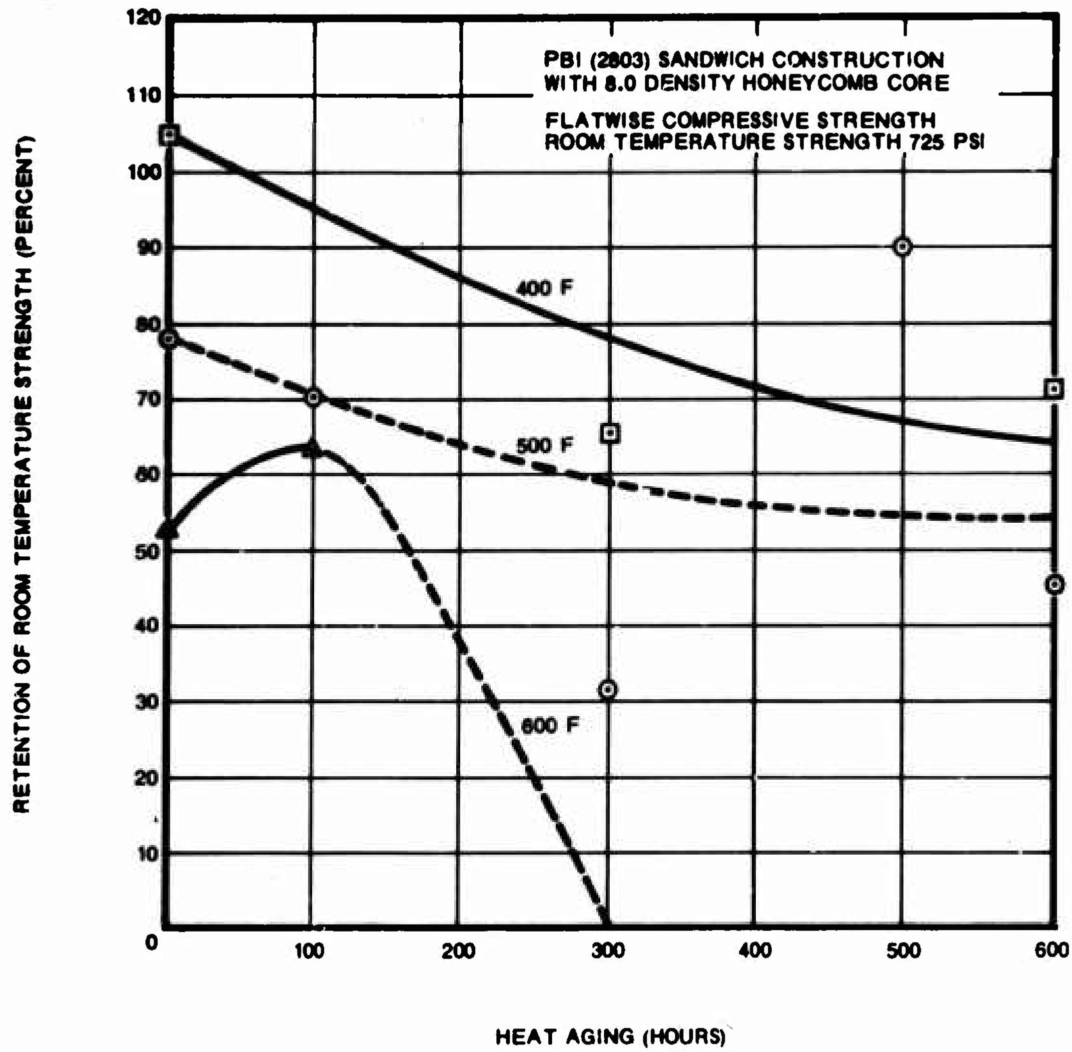



Figure 65 - PBI Composite, 8.0-lb Core, Compressive Strength, Hours Aging versus Percent Retention of Room Temperature Strength

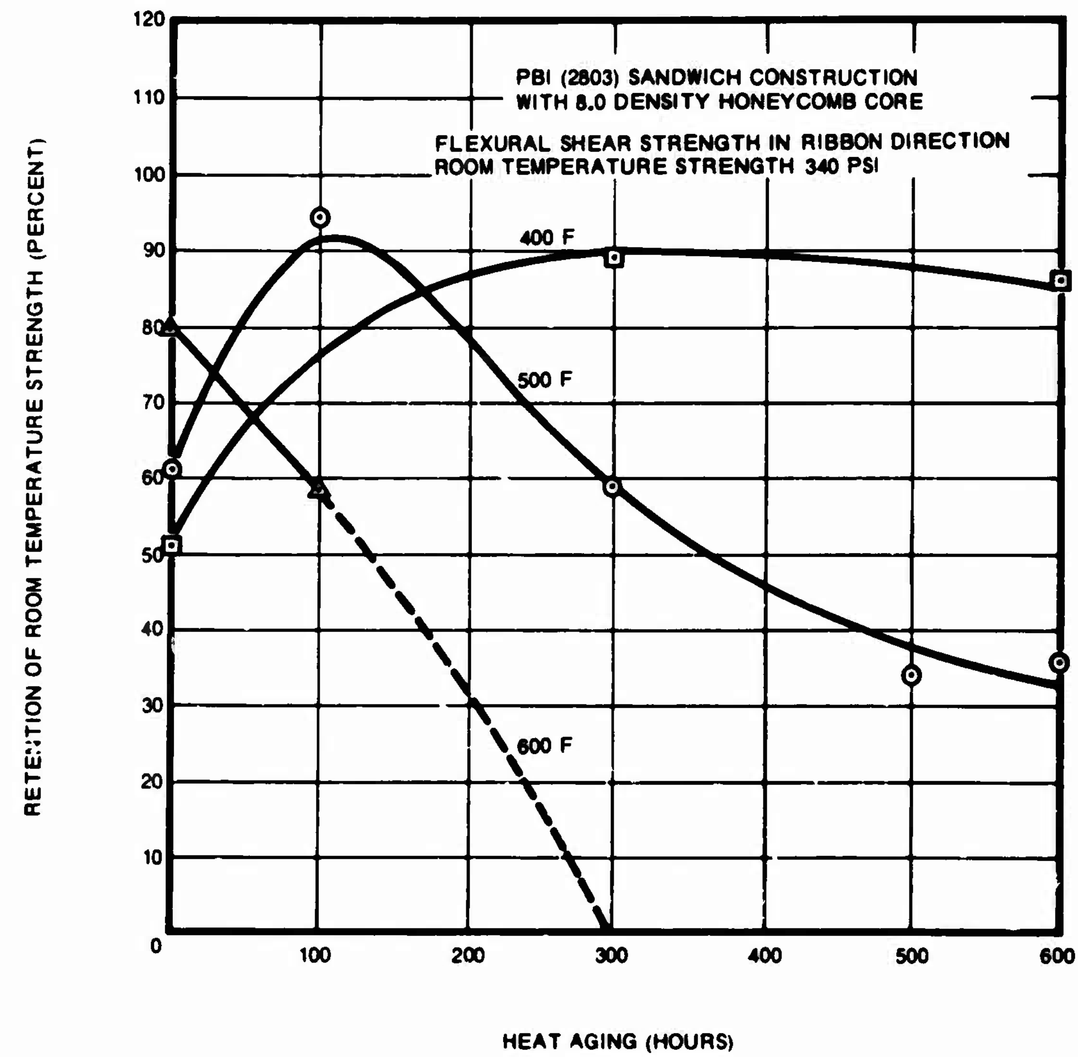



Figure 66 - PBI Composite, 8.0-lb Core, Ribbon Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

Figure 67 - Composite Bending Modulus - Ribbon Direction - 8.0 lb. Core
Figure 68 - Flexural Shear Strength - Transverse Direction - 8.0 lb. Core
Figure 69 - Composite Bending Modulus - Transverse Direction - 8.0 lb. Core

The figures present a graphical picture of the changes in the physical strengths of the PBI composites during heat aging at 400F, 500F, and 600F over a period of 600 hours.

The 600F aging curves are all dotted between the strength value at 100 hours and zero strength at 300 hours to indicate complete failure at sometime during this period.

Some of the 400F and 500F heat aging curves in Figures 58 through 69 are shown as dotted lines because test values varied to such an extent that the actual position and trend of the heat aging curves were nearly impossible to determine.

These poorly established curves which are dotted in Figures 58 through 69 are shown in Figures 70, 71, and 72 as solid lines with the actual test values marked on the graphs. The extensive scatter of the points is readily seen and the confusion this presents in attempting to draw in a representative, yet logical, curve becomes apparent. The corresponding curves at the other heat aging level (400F or 500F) are dotted in on the graphs of Figures 70, 71, and 72 to give some visible evidence as to why the shapes of the poorly established curves are presented as shown. The general trend of the PBI heat aging curves would strongly indicate that the higher the aging temperature, the lower the strength and the more rapid the loss of strength as aging time increases.

The curves which are difficult to establish based on the actual test results are presented in Figures 70, 71, and 72 in such a manner as to correspond to the general trend.

## C. DISCUSSION OF HEAT AGE DATA

1. In all cases, specimens heat-aged at 600F experienced catastrophic deterioration sometime after 100 hours. Resin burn-out, adhesive deterioration, and core degradation occurred to such an extent between 100-hours and 300-hours aging periods that the samples literally fell apart and could not be tested.

2. The heat aging results at 400F and 500F indicated that oxygen degradation became less severe at the lower temperatures. Strength values were still above 50% of room temperature

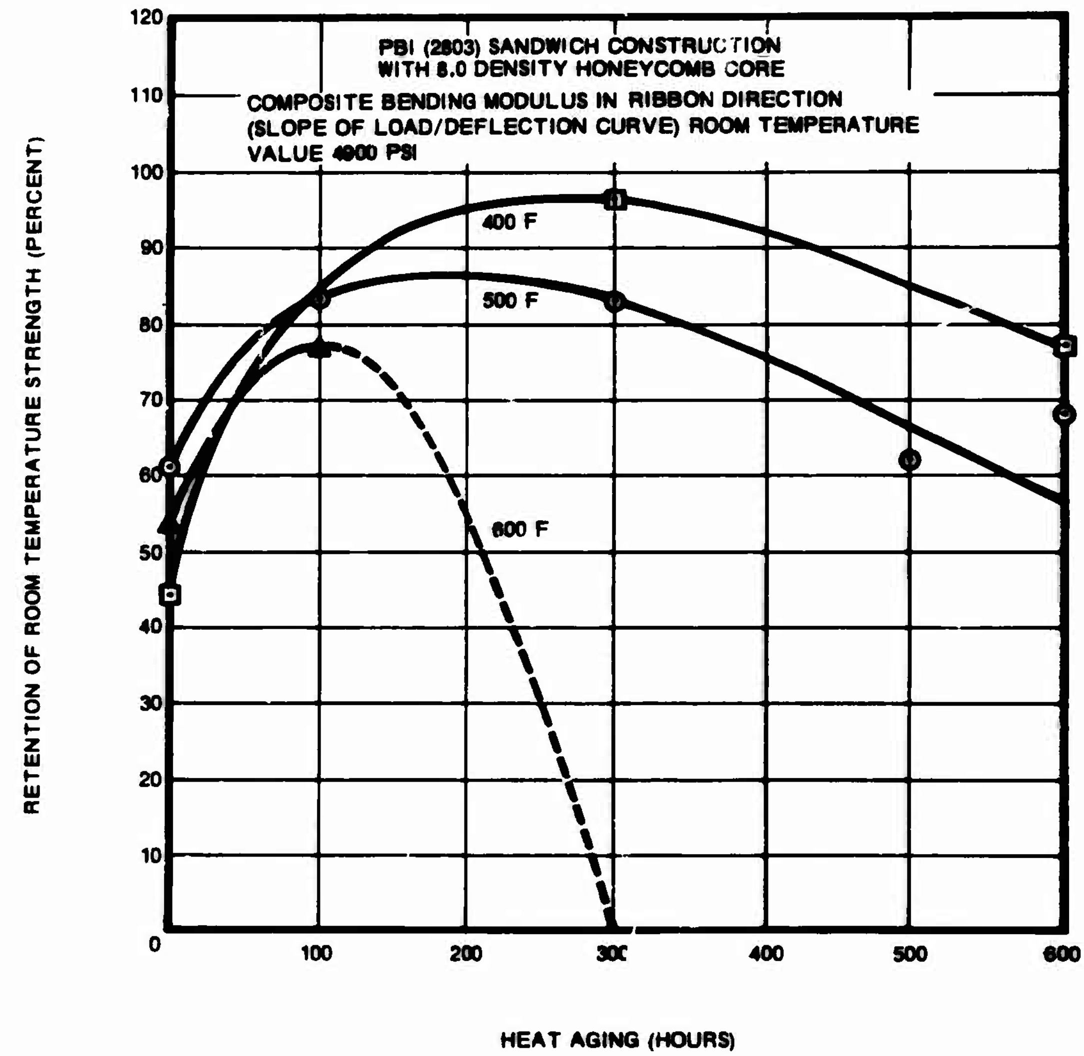



Figure 67 - PBI Composite, 8.0-lb Core, Ribbon Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength

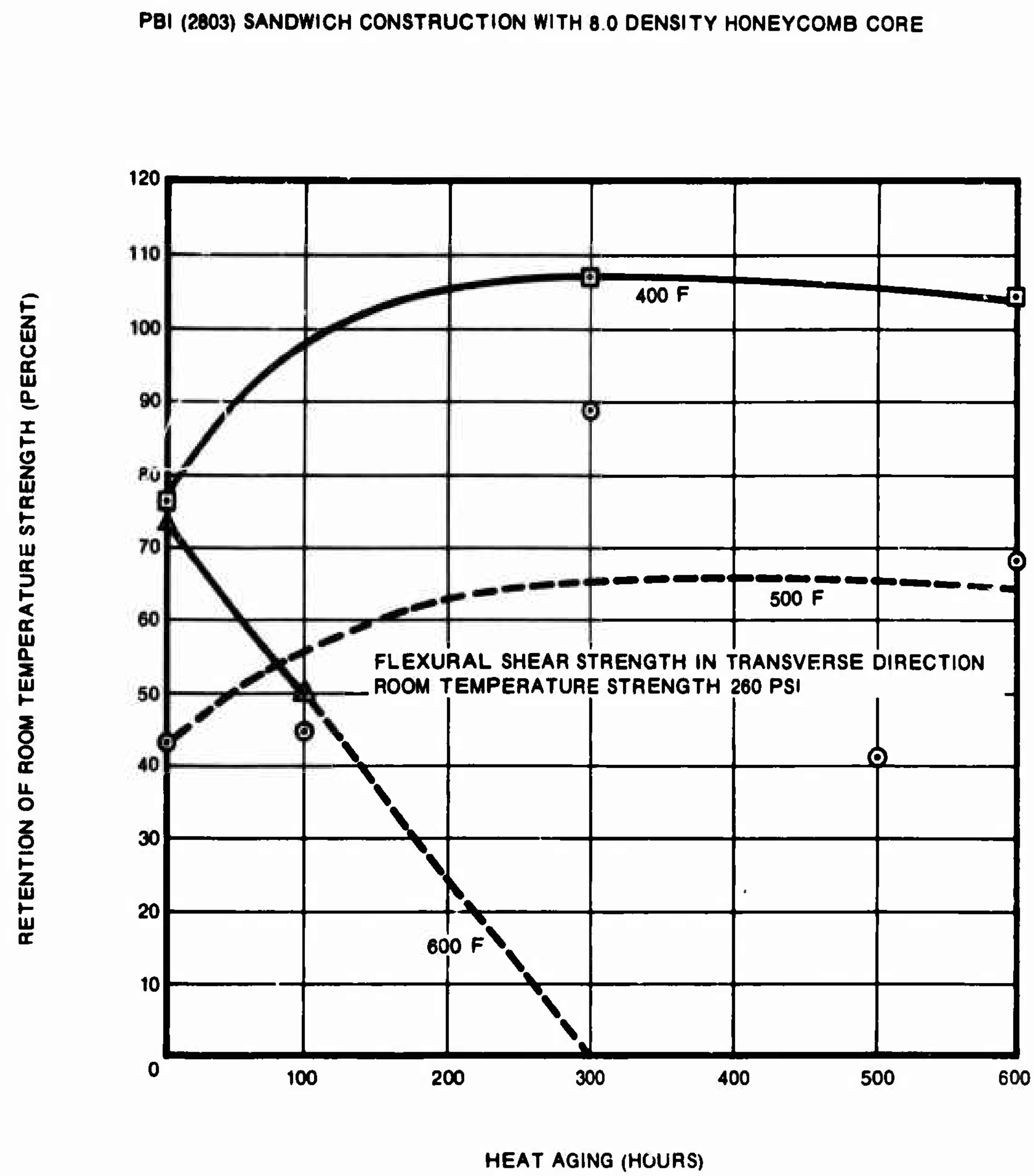



Figure 68 - PBI Composite, 8.0-lb Core, Transverse Flexural Shear Strength, Hours Aging versus Percent Retention of Room Temperature Strength

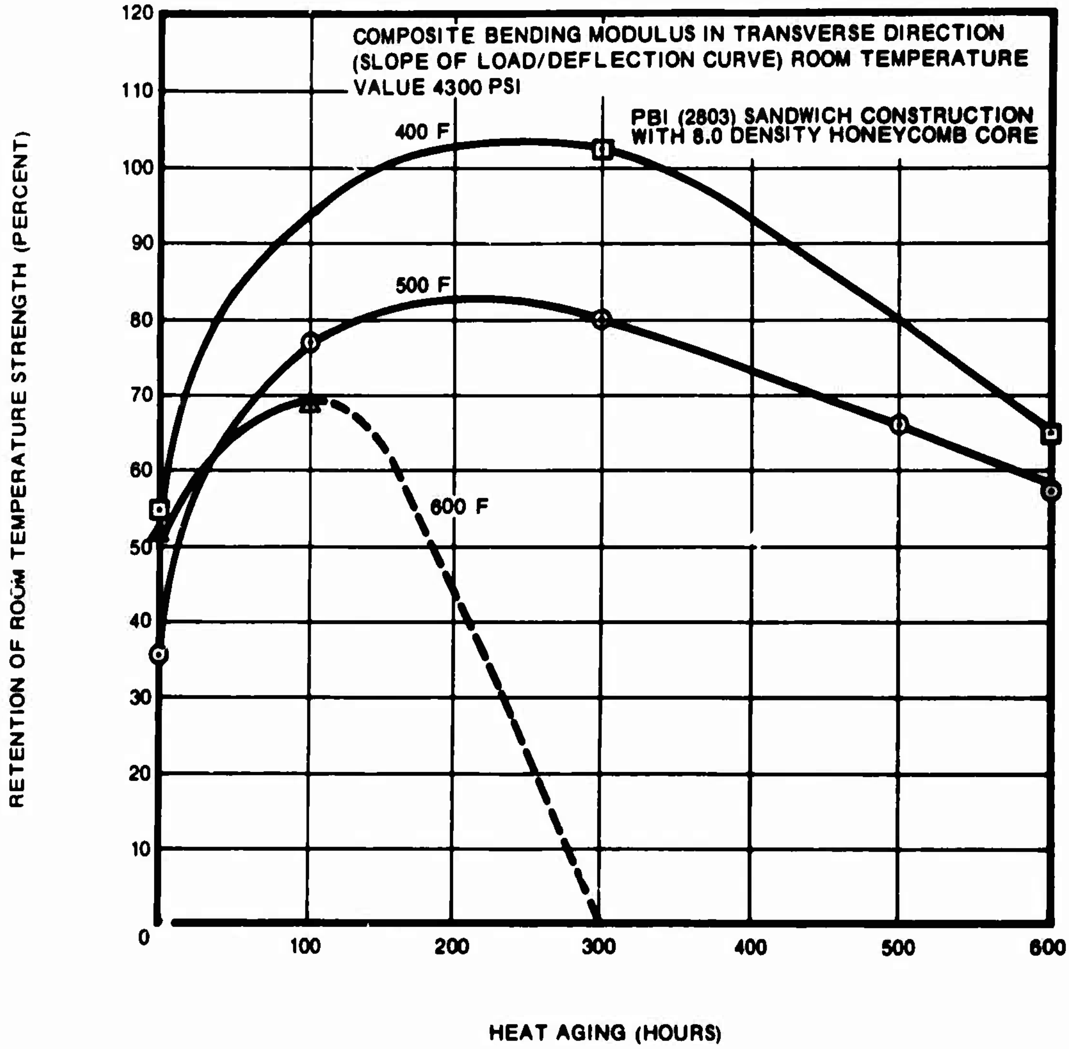



Figure 69 - PBI Composite, 8.0-lb Core, Transverse Composite Bending Modulus, Hours Aging versus Percent Retention of Room Temperature Strength

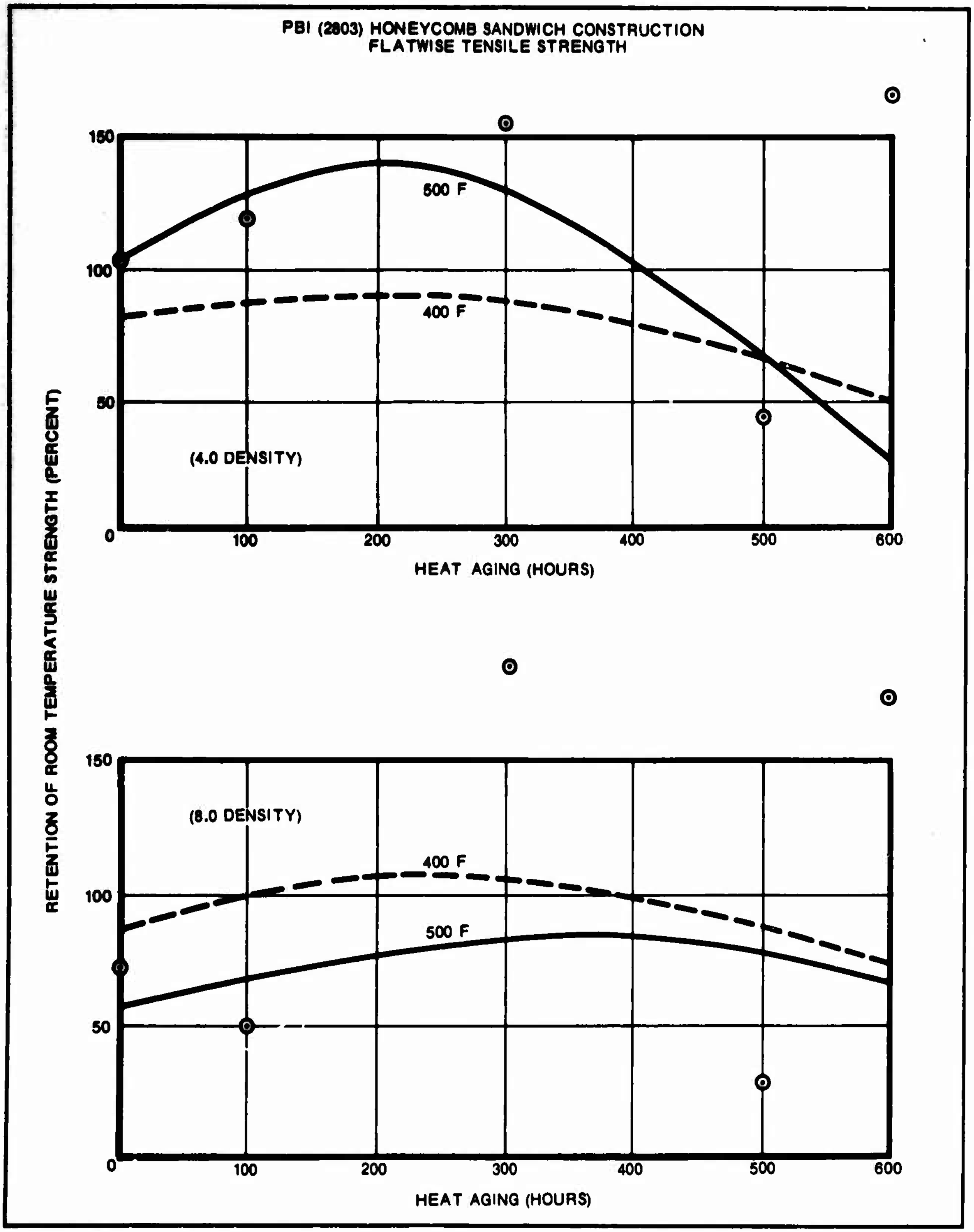



Figure 70 - PBI Composite, Tensile Strength Curves, Hours Aging versus Percent Retention of Room Temperature Strength

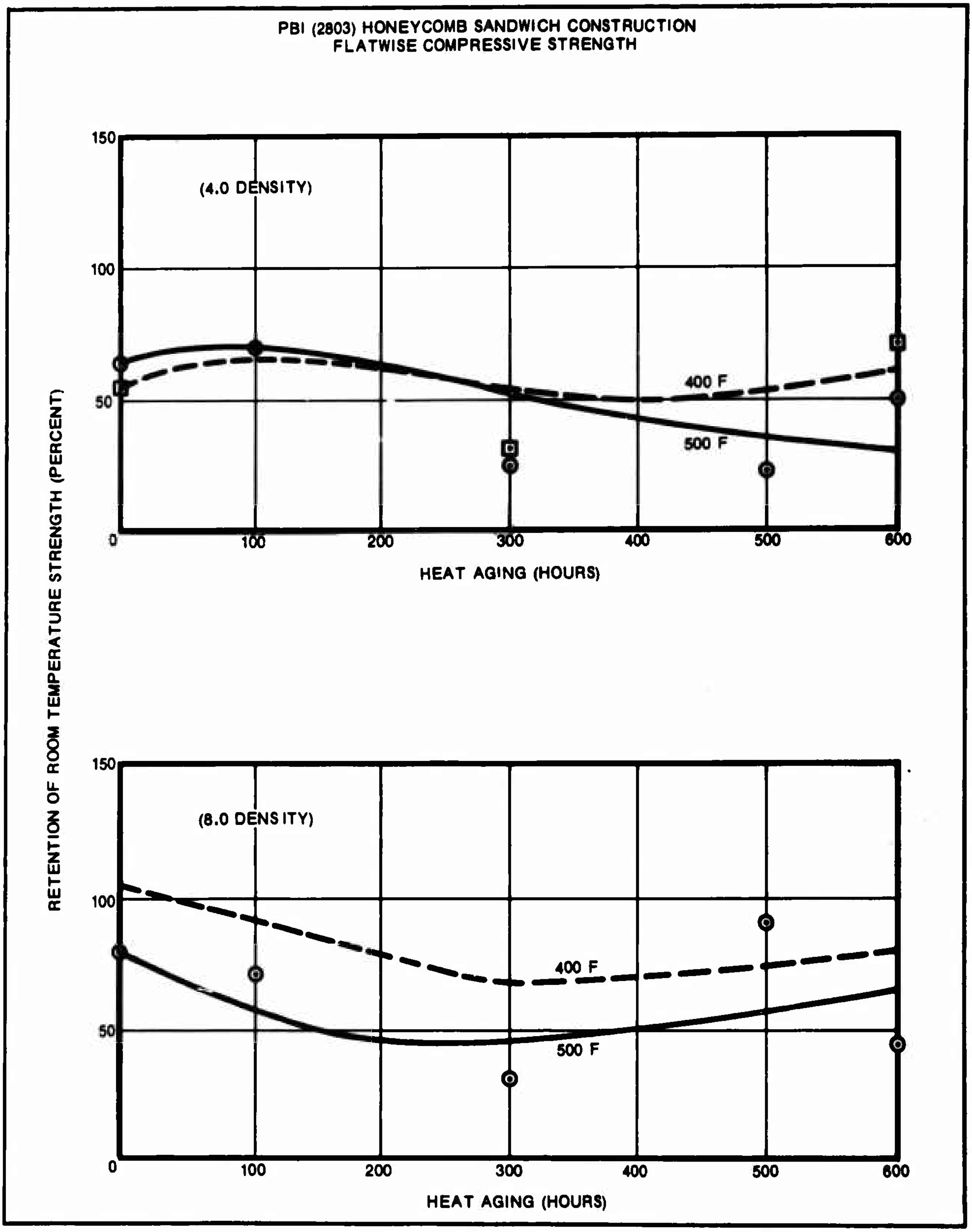



Figure 71 - PBI Composite, Compressive Strength Curves, Hours Aging versus Percent Retention of Room Temperature Strength

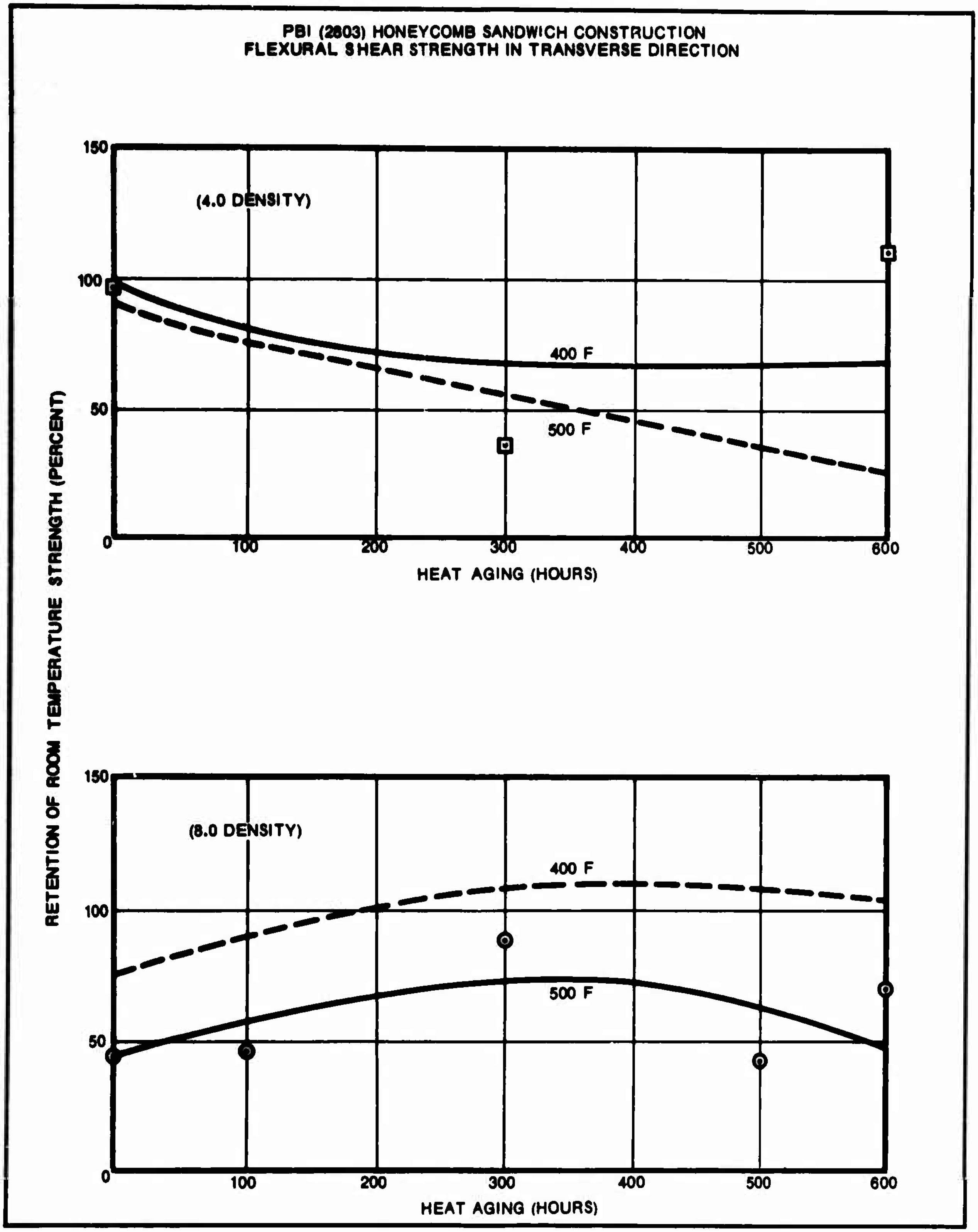



**Figure 72 - PBI Composite, Transverse Flexural Shear Strength Curves, Hours Aging versus Percent Retention of Room Temperature Strength**

values after aging for 600 hours at 400F.

3. The 8.0 lb. density core composite was stronger than the 4.0 lb. density core composite with respect to the physical properties tested. The increase in strength, however, was not very great.

4. Test results on PBI composites were varied and unpredictable. A number of reasons have been advanced for this variability:

   a. Variations in the prepreg. - The PBI (2803) prepreg was quite non-uniform. Appearance, resin content, and resin consistency varied from roll to roll, along the length of the roll, and across the roll.

   b. Variations in the honeycomb core. - The PBI (2803) honeycomb core stock was also non-uniform. Variations were apparent in resin content, porosity, and resin filleting at the node bond lines.

   c. Variations in adhesive film. - The PBI (2801) adhesive film varied in the same manner as the PBI (2803) prepreg. The fact that the prepreg contained the AFR-151 resin while the adhesive film contained the AFR-100 resin added another variable. The two PBI resins differed in reaction temperature, outgassing violence and composition, and cure temperature.

   d. Oxygen degradation during cure. - Any air leakage into the bagged composite during the autoclave cure caused resin degradation. In several cases, air contamination during cure was detected by the severe discoloration imparted to the composites. These sample parts were discarded. It is possible, however, that minor bag leakage could occur and go undetected. The PBI resin was a dark brown and discoloration due to slight resin degradation could not be seen.

   e. Oxygen degradation during post cure. - The PBI sandwich composites were post cured under a blanket of argon because of the susceptibility to oxygen degradation at the post cure temperatures. Any traces of oxygen caused resin degradation. With the honeycomb core sandwich composites, prolonged purging of the post curing chamber was necessary before the start of the post cure cycle. Air entrapment within the core cells could be a source of non-uniform degradation.

f. Oxygen degradation during heat aging. - The PBI composites were heat aged in ovens with forced air circulation. No attempt was made to protect the samples from oxygen attack. The test specimens were cut oversize for heat aging and then trimmed to exact size for testing in order to minimize edge effect. The value of this procedure was questionable, however, because of the extreme porosity of the honeycomb core. During heat aging the test specimens were supported on special racks which kept each specimen separated from the others. Even with this precaution it is possible that oxygen attack could vary due to position on the support rack and due to location within the oven.

g. Curing pressure. - It was found that the autoclave pressure had to be carefully controlled during the cure cycle to prevent core crushing. A pressure of 50 psi was tried initially but was found to be too high. Pressures of 30 psi, 25 psi, and finally 20 psi were then investigated.

The lower pressure of 20 psi was used for processing the heat age test samples when indications of core crushing were detected at higher pressures. Pressures this low caused considerable concern, especially since an aluminum foil was used for bagging and the bag was not under vacuum but vented through the autoclave wall to the atmosphere.

## SECTION XI

## CONCLUSIONS

### A. POLYIMIDE RESIN SYSTEM

1. Satisfactory thin face sheet laminates can be prepared by both press and autoclave methods.

2. A "pressure-point" cure technique was developed which provided a controlled, efficient method for fabricating face sheet laminates. The technique was applicable to both press and autoclave processing.

3. A press cure of 20 minutes at 600F under 250 PSI pressure produced four-ply face sheet laminates which did not require post curing. Flexural strengths in excess of 75,000 PSI (RT) were obtained.

4. An autoclave cure of 90 minutes at 400 F under 50 PSI pressure produced four-ply face sheet laminates which did not require post curing. Flexural strengths in excess of 75,000 PSI (RT) were obtained.

5. The use of a treated release cloth peel ply on one surface of the laminate produced an excellent bondable surface for secondary bonding operations.

6. Satisfactory sandwich composites could be prepared by secondary bonding. Acceptable secondary bonds were made by both the press and autoclave methods.

7. Single-stage fabrication and multiple-stage fabrication produced satisfactory sandwich composites in the autoclave method. The single-stage technique was most satisfactory from the standpoint of overall properties and fabrication efficiency.

8. The "pressure-point" cure technique was applicable to the autoclave fabrication of honeycomb core sandwich composites.

9. The sandwich composites with the 8.0 lb. per cu. ft. core density were stronger than the composites using the 4.0 lb. core. Physical properties were approximately proportional to core density.

10. The polyimide sandwich composites exhibited excellent resistance to heat aging.

11. The sandwich composite with the 4.0 lb. core density retained 69% of the room temperature shear strength after 1400 hours at 400F, 58% after 1400 hours at 500F; and 53% after 600 hours at 600F. Aged samples were tested at aging temperature.

12. The sandwich composite with 8.0 lb. core density retained 63% of the room temperature shear strength after 1400 hours at 400F; 61% after 1200 hours at 500F; and 42% after 600 hours at 600F. Aged samples were tested at aging temperature.

B. PBI RESIN SYSTEM

1. Thin face sheet laminates were successfully prepared by both the press and autoclave methods.

2. The "pressure-point" cure technique was used successfully for both press and autoclave fabrication of thin face sheet laminates.

3. A press cure of 90 minutes at 700F under 200 PSI pressure produced four-ply AFR-151(PBI) laminates which exhibited flexural strength values in excess of 80,000 PSI (RT). Post curing did not appear to be necessary, but was considered appropriate because of the high conversion temperature of the resin.

4. In the autoclave fabrication of AFR-151 (PBI) laminates and sandwich composites, a soft aluminum bag sealed by uncured silicone rubber tape and silicone gaskets was required to withstand the high cure temperatures.

5. In autoclave processing it was necessary to vent the part to the atmosphere.

6. Autoclave temperatures could be kept below 550 F provided an extended 5-1/2-hour step-cure cycle was employed.

7. The cure cycle could be shortened to four hours by using the pressure-point technique and an autoclave temperature of 620F.

8. Face sheet laminates prepared by the autoclave method required a 34-hour post cure treatment. Flexural strength values of four ply laminates were in excess of 80,000 PSI (RT).

9. The use of a treated release cloth peel ply produced a satisfactory bondable surface for secondary bonding.

10. Satisfactory sandwich composites were prepared by secondary bonding using both press and autoclave methods.

11. Satisfactory honeycomb core sandwich composites were prepared in the autoclave using single-stage and multiple stage fabrication methods. The single stage method was considered best.

12. The "pressure-point" cure technique was applied successfully to the autoclave fabrication of PBI sandwich composites.

13. The AFR-151 (PBI) resin system was highly susceptible to oxidation during cure and post cure.

14. The strength values of the PBI sandwich composites were variable and unpredictable. This was felt due to the excessive variations encountered in supplied materials, the extreme processing conditions; and oxygen degradation.

15. The PBI sandwich construction was subject to oxygen degradation during heat aging. Degradation increased as aging temperature increased. Test specimens underwent catastrophic failure in less than 300 hours at 600F.

16. The sandwich composite with 4.0 lb. core density retained 84% of the room temperature shear strength after 600 hours at 400F; 45% after 600 hours at 500F. Aged samples were tested at aging temperature.

17. The sandwich composite with 8.0 lb. core density retained 86% of the room temperature shear strength after 600 hours at 400F; 36% after 600 hours at 500F. Aged samples were tested at aging temperature.

## SECTION XII

## RECOMMENDATIONS

1. The static heat aging test program started in this project should be continued until strength values drop below the 50% level, or for a minimum of 10,000 hours. This would provide the long-time heat aging data felt necessary for design of supersonic aircraft components with a high level of confidence.

2. The polyimide resin sandwich composite system should be subjected to a more rigorous test program. Fatigue testing, cyclic testing, and heat aging under load should be included to provide test conditions approaching the actual environment of future supersonic aerospace vehicles.

3. New improved sandwich core constructions using polyimide resin as the binder should be evaluated.

4. Metal-plastic structural sandwich constructions should be studied. Titanium-polyimide combinations should definitely be included. Processing studies and heat age testing should be performed.

5. Studies should be developed for reducing the susceptibility of PBI sandwich constructions to oxygen degradation. These studies should include an evaluation of sealants, barrier films, and core fillers. Process improvement techniques should also be explored.

Unclassified
Security Classification

# DOCUMENT CONTROL DATA - R&D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

| | |
|---|---|
| 1. ORIGINATING ACTIVITY (Corporate author)<br>Goodyear Aerospace Corporation<br>Litchfield Park, Arizona | 2a. REPORT SECURITY CLASSIFICATION<br>Unclassified<br>2b. GROUP |

3. REPORT TITLE

Fabrication and Test of High-Temperature Honeycomb Core Sandwich Construction

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Final: 1 June 1966 through 30 June 1967

5. AUTHOR(S) (Last name, first name, initial)

Wintermute, G. E.

| 6. REPORT DATE | 7a. TOTAL NO. OF PAGES | 7b. NO. OF REFS |
|---|---|---|
| September 1967 | 182 | |

| | |
|---|---|
| 8a. CONTRACT OR GRANT NO.<br>AF33(615)-5131<br>b. PROJECT NO.<br>7381, Task No. 738101<br>c.<br>d. | 9a. ORIGINATOR'S REPORT NUMBER(S)<br>AFML-TR-67-273<br>9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report) |

10. AVAILABILITY/LIMITATION NOTICES

This document is subject to special controls and each transmittal to foreign governments or foreign nationals may be made only with prior approval of the Air Force Materials Laboratory, MAAE, Wright-Patterson Air Force Base, Ohio, 45433.

| | |
|---|---|
| 11. SUPPLEMENTARY NOTES | 12. SPONSORING MILITARY ACTIVITY<br>AFML (MAAE)<br>Wright-Patterson Air Force Base, Ohio |

13. ABSTRACT


The primary objective of this program was the determination of the engineering properties of polybenzimidazole and polyimide honeycomb core sandwich constructions after aging at various temperatures.

The work was conducted in three phases: I. Processing and Materials Study; II. Heat Aging and Testing; III. Data Correlation.

Both press and autoclave methods were evaluated for the fabrication of sandwich face sheets and honeycomb core sandwich composites. A "pressure-point" cure technique was developed which provided a controlled, efficient processing method for preparing polyimide and PBI laminates and sandwich composites.

Technology was developed for fabricating sandwich composites by secondary bond, single stage cure, and multiple stage cure methods. The single stage cure method, using the pressure-point technique, was selected for preparing the heat aging test specimens for both the polyimide and the PBI resin systems.

Heat aging studies were conducted at 400F, 500F, and 600F. Polyimide sandwich composites exhibited excellent heat resistance at all test temperatures. The polyimide composites retained over 50% of their room temperature flexural shear strength values after 1400 hours aging at 400F.

The PBI sandwich composites were susceptible to oxygen degradation during fabrication and during the heat aging study. Specimens heat aged at 600F exhibited complete failure within a 300-hour period. Oxygen attack was less at lower temperatures and PBI test specimens still retained over 50% of their room temperature strength values after 600 hours aging at 400F.


DD FORM 1 JAN 64 1473

Unclassified

Unclassified
Security Classification

| 14. KEY WORDS | LINK A | | LINK B | | LINK C | |
|---|---|---|---|---|---|---|
| | ROLE | WT | ROLE | WT | ROLE | WT |
| Polyimide<br>Polybenzimidazole<br>Honeycomb core sandwich structure<br>Composite<br>Heat aging<br>Mechanical properties<br>Processing - press<br>Processing - autoclave | | | | | | |

## INSTRUCTIONS

1. ORIGINATING ACTIVITY: Enter the name and address of the contractor, subcontractor, grantee, Department of Defense activity or other organization *(corporate author)* issuing the report.

2*a*. REPORT SECURITY CLASSIFICATION: Enter the overall security classification of the report. Indicate whether "Restricted Data" is included. Marking is to be in accordance with appropriate security regulations.

2*b*. GROUP: Automatic downgrading is specified in DoD Directive 5200.10 and Armed Forces Industrial Manual. Enter the group number. Also, when applicable, show that optional markings have been used for Group 3 and Group 4 as authorized.

3. REPORT TITLE: Enter the complete report title in all capital letters. Titles in all cases should be unclassified. If a meaningful title cannot be selected without classification, show title classification in all capitals in parenthesis immediately following the title.

4. DESCRIPTIVE NOTES: If appropriate, enter the type of report, e.g., interim, progress, summary, annual, or final. Give the inclusive dates when a specific reporting period is covered.

5. AUTHOR(S): Enter the name(s) of author(s) as shown on or in the report. Enter last name, first name, middle initial. If military, show rank and branch of service. The name of the principal author is an absolute minimum requirement.

6. REPORT DATE: Enter the date of the report as day, month, year; or month, year. If more than one date appears on the report, use date of publication.

7*a*. TOTAL NUMBER OF PAGES: The total page count should follow normal pagination procedures, i.e., enter the number of pages containing information.

7*b*. NUMBER OF REFERENCES: Enter the total number of references cited in the report.

8*a*. CONTRACT OR GRANT NUMBER: If appropriate, enter the applicable number of the contract or grant under which the report was written.

8*b*, 8*c*, & 8*d*. PROJECT NUMBER: Enter the appropriate military department identification, such as project number, subproject number, system numbers, task number, etc.

9*a*. ORIGINATOR'S REPORT NUMBER(S): Enter the official report number by which the document will be identified and controlled by the originating activity. This number must be unique to this report.

9*b*. OTHER REPORT NUMBER(S): If the report has been assigned any other report numbers *(either by the originator or by the sponsor)*, also enter this number(s).

10. AVAILABILITY/LIMITATION NOTICES: Enter any limitations on further dissemination of the report, other than those imposed by security classification, using standard statements such as:

(1) "Qualified requesters may obtain copies of this report from DDC."

(2) "Foreign announcement and dissemination of this report by DDC is not authorized."

(3) "U. S. Government agencies may obtain copies of this report directly from DDC. Other qualified DDC users shall request through
_______________________________."

(4) "U. S. military agencies may obtain copies of this report directly from DDC. Other qualified users shall request through
_______________________________."

(5) "All distribution of this report is controlled. Qualified DDC users shall request through
_______________________________."

If the report has been furnished to the Office of Technical Services, Department of Commerce, for sale to the public, indicate this fact and enter the price, if known.

11. SUPPLEMENTARY NOTES: Use for additional explanatory notes.

12. SPONSORING MILITARY ACTIVITY: Enter the name of the departmental project office or laboratory sponsoring *(paying for)* the research and development. Include address.

13. ABSTRACT: Enter an abstract giving a brief and factual summary of the document indicative of the report, even though it may also appear elsewhere in the body of the technical report. If additional space is required, a continuation sheet shall be attached.

It is highly desirable that the abstract of classified reports be unclassified. Each paragraph of the abstract shall end with an indication of the military security classification of the information in the paragraph, represented as *(TS)*, *(S)*, *(C)*, or *(U)*.

There is no limitation on the length of the abstract. However, the suggested length is from 150 to 225 words.

14. KEY WORDS: Key words are technically meaningful terms or short phrases that characterize a report and may be used as index entries for cataloging the report. Key words must be selected so that no security classification is required. Identifiers, such as equipment model designation, trade name, military project code name, geographic location, may be used as key words but will be followed by an indication of technical context. The assignment of links, rules, and weights is optional.

Unclassified
Security Classification